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JNMM Journal of Nuclear Materials Management

National Nuclear Forensics Library Virtual Tabletop Exercise "Galaxy Serpent"

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Project ID: 1
Configuration: 2
Matrix: C
Container:
Analysis Date: 6/7/2012 11:00:30 AM
Acquisition Date: 6/6/2012 11:29:28 AM
Scan ID: Pu Capsule
Detector: Dspec ep44

Isotope(iPeak)	Activity	Weight	Error	Total Error	Matrix Error
Pu-238	25.6876E+07	4.14	0	.00	.00
NP-237	25.5759E+06	.01	0.1007	47.88	.00
PU-241	17.4990E+02	.00	1.1091	47.90	.00
AM-241	30.0955E+08	.03			
PU-239	.0000E+00	.00			
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INMM
INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT

Facing Our Challenges

By Ken Sorenson
INMM President

I hope everyone is having a great summer and is paying attention to the right mix of business and pleasure in your lives. Of course, I hope part of this mix is the 55th INMM Annual Meeting! As this is my last column, the traditional focus would be on reflections. However, I would like to use this space to juxtapose the recent challenges for the INMM concerning U.S. government sponsored attendance at our meetings with the importance of the global INMM mission.

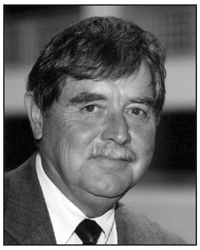
Two years ago, the U.S. government applied strict conference attendance criteria to individuals sponsored by U.S. government programs and using U.S. government funds. Interpretation of guidelines and application of the guidance was uneven across U.S. government agencies. The U.S. Department of Energy (DOE), one of INMM's main U.S. government participants, took a strict interpretation of the guidance and severely cut back on the number of participants from DOE-sponsored programs that were approved to attend annual meetings. The impact on the INMM has been significant. Annual meeting attendance has gone from a high in 2011 of 1,167, to 970 in 2012, and 594 in 2013. This type of precipitous decline reflects more than just the number of U.S. participants. We also saw a sharp decline in the number of non-U.S. participants who are deeply involved in U.S. nonproliferation activities, as well as the number of students sponsored by U.S. DOE programs. The financial impact of these reduced numbers has resulted in a sharp decline in our annual operating revenues. Balancing these reduced revenues with operation costs has been a particular challenge. Since recent hotel commitments are set years in advance and attendance projections are based on past performance (i.e., on strong attendance figures leading up to and including 2011), we have had a difficult couple of years working with the host hotels to negotiate contract terms based on the reduced registrations over the past two years.

The 55th Annual Meeting reflects, in my opinion, a shifting of the downward trend to a positive direction. I believe this change in direction is driven in large part by the importance of the INMM mission (but, I'll get to that later!). For the 55th Annual Meeting, we are projecting to have a little more than 700 registrants. Additionally, the number of approved DOE registrants is 242. This number is up 66 from 2013, a nearly 40 percent increase. Student paper abstracts total 130, a record high. With these projections, we anticipate a net surplus in our FY14 financial operations, a huge turnaround from FY13.

This upward trend points to the important mission space that INMM fills. First, there is recognition within the DOE that the INMM Annual Meeting and workshops sponsored by the INMM align well with important mission spaces of the DOE. This has resulted in an expansion of the number of approved DOE-sponsored registrants at the INMM Annual Meeting this year. Second, the INMM is still experiencing an expansion of regional and student chapters around the globe. In particular, we are seeing this expansion in new regions that understand the importance of the INMM mission and want to be a part of this global effort. We have approved new regional chapters in Morocco and Nigeria as well as student chapters in Jordan and India. Third, our formal relationships with external organizations are strong and provide important cross-fertilization functions. Specifically, we have formal agreements with the World Institute of Nuclear Security, the European Safeguards Research and Development Association, and the Nuclear Infrastructure Council. With all of these organizations, we are active in sponsoring joint workshops related to specific technical areas in safeguards, security, safety, and operations.

In the current "*Taking the Long View in a Time of Great Uncertainty*" article in this issue, Jack Jekowski outlines five challenges for the next generation of nuclear stewards. You will notice that these five challenges relate directly to the INMM mission.

The INMM continues to lead in the international effort to manage nuclear materials in a safe, secure, and responsible manner.



Galaxy Serpent: A Special Issue on National Nuclear Forensics Libraries

By Dennis Mangan

On June 11, 2013, I received an e-mail from a dear friend, Bill Severe, who was working at that time in the U.S. Department of State's Office of Weapons of Mass Destruction Terrorism. This office was supporting activities of the Nuclear Forensics International Technical Working Group, which had conducted a web-based exercise called Galaxy Serpent, involving about twenty countries and that was aimed at improving skills in identifying the origin of seized nuclear materials. As Severe noted, "We would like to give the participating countries the opportunity to have their experience and technical findings published in a single technical publication and the *Journal* seems to be an ideal publication for that purpose." To get things started and planned, Dr. Jim Borgardt of the State Department organized a conference call involving the *JNMM* editors and appropriate State Department and U.S. Department of Homeland Security personnel. Frank Wong of the U.S. Department of Homeland Security was likewise instrumental in the planning that occurred. The decision was made to have about six articles from different countries available for this issue of the *Journal*. As it turned out, nine countries/organizations provided articles: Australia, Brazil, Canada, Japan, South Africa, the United Kingdom, Hungary, Sweden, and the European Commission (Joint Research Centre). Drs. Borgardt and Wong provide the "introductory" article to the topic addressed in the remaining nine articles. This issue on developing a national nuclear forensics library is definitely interesting. I trust you will enjoy reading the articles by the nine countries/organizations.

Regarding Regular Features

In his message, our President Ken Sorenson expresses a positive attitude regarding the 55th INMM Annual Meeting. As he states, "The 55th Annual Meeting reflects, in my opinion, a shifting of the downward trend to a positive direction." Those are good words to read.

Book Review Editor Mark Maiello provides an interesting review of *A Global History of the Nuclear Arms Race.* The book is divided into two volumes, one covering the 1930s to the 1980s, and the other from the 1980s to the present day. As usual, Mark presents a detailed review that makes one want to go out and get the book.

Jack Jekowski, chair of the INMM Strategic Planning Committee and editor of *Taking the Long View,* has an excellent article titled "Throwing Down the Gauntlet to the Next Generation of Nuclear Stewards — the Enduring Nuclear Legacy." His article has much history in it and is enjoyable to read. This is not unusual for Jack.

The Bonus of Digital Publishing

This is the end of the first full publishing year as a digital publication. We have worked to make the transition smooth for everyone involved, especially our readers. We have provided bonus content with every issue, ranging from articles that we posted only on the INMM website to larger-sized images from some of our articles. With this issue, we are introducing a permanent "bonus" for *JNMM* readers: a glossary of abbreviations and acronyms commonly used in the *Journal*. We plan to update this list with each issue of *JNMM*, creating an easy reference that you can use.

I trust you will enjoy this issue of the *JNMM*. We owe much thanks to those who initiated and accomplished this special issue on the topic of national nuclear forensics libraries.

JNMM *Technical Editor Dennis Mangan can be reached at dennismangan@comcast.net*

Galaxy Serpent: A Web-based Tabletop Exercise Using the Concept of National Nuclear Forensics Libraries

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Abstract

Galaxy Serpent is a first-of-a-kind, virtual, web-based international tabletop exercise where teams of scientists from various countries used provided public domain spent fuel compositions to formulate their own national nuclear forensics library (NNFL), and determined if hypothetically seized spent nuclear fuel is or is not consistent with their national nuclear forensics library. This tabletop exercise is conducted under the auspices of the Nuclear Forensics International Technical Working Group (ITWG) and funded and organized by the U.S. Department of State with technical expertise provided by the U.S. Department of Homeland Security. It involved scientists from approximately twenty-four countries, and the active participation of eighteen teams. *Galaxy Serpent* aims to promote "best practices" through providing a vehicle for participants to gather key technical expertise to create an NNFL using guidelines in International Atomic Energy Agency (IAEA) documents and to illustrate the potential probative benefits offered by creating such a library. During the play of *Galaxy Serpent*, many teams quickly realized the need to involve other areas of expertise such as nuclear reactor engineers and fuel experts. The involvement of such additional experts helps to mature the range of expertise of the nuclear forensics international community. Teams also noted that different technical approaches yielded similar analytical conclusions, and that the original purpose, history, and limitations of the provided data sets sometimes limited the confidence levels associated with their findings. In addition, some of *Galaxy Serpent* teams have used this tabletop exercise experience to inform their efforts at home to develop or refine their own NNFLs. The exercise has yielded insightful lessons regarding the efficacy NNFLs can provide, and how they, in concert with traditional forensics, may expand the suite of tools for investigating nuclear or other radioactive material outside legitimate control.

Introduction

Several high-profile law enforcement seizures in the mid-1990s demonstrated the availability illicit nuclear or other radioactive (RN) material on the black market was increasing. In 1995 the International Atomic Energy Agency (IAEA) established what is now called the Incident and Trafficking Database (ITDB), a voluntary program to report incidents of illicit trafficking and other unauthorized activities and events involving RN materials outside of regulatory control. Between January 1993 and December 2012, a total of 2,331 incidents were reported to the ITDB, including 419 that involved unauthorized possession and related criminal activities such as movement or attempts to illegally trade or use nuclear material or radioactive sources. Sixteen reported incidents in this category involved weapons grade uranium or plutonium, some in kilogram quantities.[1]

Recent cases demonstrate the continued availability of nuclear and other radioactive material on the black market despite the significant progress that has been made in securing such materials. These incidents have motivated the international scientific and policy communities to develop stronger investigative tools that have proven so effective in removing RN materials from the black market. The scientific community has contributed significantly to this effort, e.g., in the area of nuclear forensics by conducting material analysis exercises, international meetings and trainings, and the development and dissemination of "best practices." This work presents a brief history of the field of nuclear forensics and recent material analysis exercises convened by a leading organization in the discipline, and introduces the first-of-a-kind virtual, web-based international nuclear forensics exercise, *Galaxy Serpent*, which advances the concept of NNFLs. Findings and lessons learned from this technical tabletop exercise (TTX) are presented.

Nuclear Forensics

Nuclear forensic science, often referred to as simply nuclear forensics, is defined as "the examination of nuclear or other radioactive material, or of other evidence that is contaminated with radioactive material, in the context of legal proceedings, including national or international law or nuclear security."[2] Nuclear forensics (NF) is an essential component of national and international nuclear security response plans to events involving nuclear and other RN out of regulatory control. The ability to collect and preserve seized RN material as evidence and conduct NF analysis may provide information about the history and origin of material, point of diversion, and identity of the perpetrators. NF is a technical capability that will also inform the investigatory process.[2] NNFLs augment these capabilities by providing an organized set of data and potential sample archives that allow comparison of illicitly trafficked material to national holdings to help determine if seized material originated in a particular country.

Nuclear forensics has become a tool to aid in identifying where loss of regulatory control may have occurred. International recognition of the danger posed by nuclear smuggling led to the development of a number of initiatives, and the formation of skill-specific groups tasked to identify and move towards effective countermeasures. For example,

- Nuclear Security Summits (NSS): Convened 2010-2014, the summits have provided effective political momentum to sustain current international collaborations in nuclear forensics per the Nuclear Security Summit Work Plan.[3]
- Global Initiative to Combat Nuclear Terrorism (GICNT):[4] An international partnership of nations that are committed to implementing a set of shared nuclear security principles that includes promoting nuclear forensics policy development, best practices, capability awareness, and tabletop exercises.
- The IAEA:[5] The IAEA develops and conducts NF guidance and training and produces Nuclear Security Series documents.
- The Nuclear Forensics International Technical Working Group (ITWG):[6] Initiated in 1995, ITWG is a volunteer consortium of international scientific practitioners serving as a technical forum to promote information exchange and analytical exercises such as the *Galaxy Serpent* tabletop exercise (TTX) reported here and comparative material analysis exercises noted below.

Nuclear Forensics International Technical Working Group (ITWG)

The *Galaxy Serpent* exercise was conducted under the auspices of the National Nuclear Forensics Libraries Task Group of the ITWG and funded and organized by the U.S. Department of State with technical expertise provided by the U.S. Department of Homeland Security. The ITWG is a multinational, informal association of official practitioners of nuclear forensics — laboratory scientists, law enforcement personnel, and regulatory officials — who share a common task in responding to nuclear security events involving nuclear or other radioactive materials out of regulatory control. The ITWG conducts its work through a combination of annual meetings, task group activities, and special exercises. Participation in the ITWG is voluntary and open to competent and qualified government participants from nations having, or wishing to have, a nuclear forensics capability.[2]

The ITWG provides a forum for the conduct of technical exercises, in which laboratories that elect to participate, can evaluate their ability to analyze nuclear material, and can compare their methods with those of the other participating laboratories. One feature of these exercises is that results are coded such that they cannot be attributed to any specific participant. This feature affords the participants a measure of anonymity as well as protection against the misapplication of the results for purposes of grading the performance of any one laboratory or any group of laboratories. Although the exercise results are anonymous, they can be used to inform next steps in NF capability development, such as enhancements in analysis techniques, availability of reference materials, and improvements in instrumentation. To date, the ITWG Exercises Task Group has overseen the development, conduct, comparative data analysis, and reporting for three collaborative material exercises (CMX).

Galaxy Serpent

Galaxy Serpent was designed with the goal of raising awareness about the technical aspects of creating and using national nuclear forensics libraries via a cost-effective, wholly web-

Table 1. Teams involved in each round

Round 1 teams (February-April 2013)	Round 2 teams (May-July 2013)	Round 3 teams (August-October 2013)	Round 4 teams (February-April 2014)	Round 5 teams (February-April 2014)
Australia/ANSTO	*Japan*	Hungary	Team 13	*JRC/ITU**
Brazil	South Africa/NECSA	*Sweden*	Team 14	Team 17
Canada	UK/AWE	Team 11	Team 15	Team 18
Team 4	Team 8	Team 12		

Italicized text indicates the team in each round that was assigned the reactor that was the source of the hypothetical seizure.
*JRC/ITU is the European Commission Joint Research Centre Institute for Transuranium Elements

based, platform. The virtual web-based format eliminated the steps of securing and shipping nuclear or other radioactive materials. A virtual exercise afforded a means of exercising national capabilities for analyzing complex data and rendering conclusions regarding these data without having to perform material characterization in a laboratory. Such considerations motivated the development of a wholly web-based, technical, tabletop exercise using public domain nuclear materials data that would focus on developing a national nuclear forensics library without requiring laboratory measurements and would also engage a broader diversity of teams and technical experts while maturing the concept of NNFLs and illustrating their potential efficacy.

The *Galaxy Serpent* exercise was proposed at the ITWG-17 annual meeting, held in June 2012 at The Hague, where teams representing participants from sixteen countries volunteered to participate. It is important to reiterate that while participants have varying nationalities, ITWG is a volunteer organization and any individual or team participation should not be construed to imply official sanction of the effort. Thus far, the TTX has involved observers and participants from twenty-four countries, including teams from eighteen countries who have actively participated in the five rounds of the exercise as of April 2014. Rounds 1-3 occurred between January 2013 and October 2013; rounds 4 and 5 concluded in April 2014. Nine teams that participated in the exercise authored articles presenting their experiences, findings, and lessons learned that compose this special issue of the *Journal of Nuclear Materials Management (JNMM)*. Table 1 lists the teams involved in each round and specifically identifies those that contributed articles. Each round was composed of three to four teams, conducted over approximately eight to ten weeks, and was identical in exercise structure and tasks posed. The articles that follow appear chronologically ordered by round, and by alphabetical order within each round.

The objectives of the *Galaxy Serpent* TTX are to have participants organize a model NNFL using provided spent fuel characteristics from three nuclear reactors ("Phase 1") and then determine if data from a hypothetical seizure of spent fuel is or is not consistent with a reactor in their model NNFL ("Phase 2"). In Phase 1 of the exercise, teams were provided existing, public domain, data sets from Spent Fuel Isotopic Composition (SFCOMPO), a database of isotopic measurements of spent fuel.[7] SFCOMPO is data collected from public domain, published literature of isotopic compositions of spent nuclear fuels (SNF) obtained through post-irradiation experiments (PIE), which are used in the validation of burnup credit methodologies. SFCOMPO consists of SNF isotopic compositions for fourteen commercial nuclear reactors in four countries (Germany, Italy, Japan, and the United States). SFCOMPO was initially developed by the Japan Atomic Energy Research Institute (JAERI), and in 2002 the database was transferred to the Organization for Cooperation and Economic Development/Nuclear Energy Agency (OECD/NEA).

The SFCOMPO database includes spent fuel data exclusively from light water reactors — seven pressurized water reactors (PWR) and seven boiling water reactors (BWR). In total, data from 246 samples are available. The database provides the composition for U, Pu, Am, Cm, and several fission products (such as Nd, Cs, and Sr). Since not only measured data but also ratios of the measured data are included, the database contains more than 10,000 data points.

While referred to as a database, SFCOMPO is actually a collection of tabular data arranged in spreadsheet form containing actual post-irradiation examination (PIE) data of SNF. In general, each SFCOMPO data set includes twelve parameters including, for instance, information identifying the specific reactor, assembly, and fuel rod position, as well as sampling position along the fuel rod. For each sample, material characteristics such as initial enrichment, isotopic concentrations, and burnup measurements, among others, may be given. Since the analyses were performed at laboratories in several countries, data sets often vary in the specific data they contain.

Exercise Artificialities and Adjustments

For use in the *Galaxy Serpent* exercise, some modifications and adjustments to the original SFCOMPO data sets had to be made due to the desire to re-purpose the SFCOMPO database for a nuclear forensics tabletop exercise. The primary artificiality is that SFCOMPO data, in its original form, is difficult to use for reactor identification as there are no uncertainties associated with the data. Since measurements would not exactly match any one particular set of reactor fuel data, without explicit data uncertainties it would be impossible to determine if the measurements for any one fuel would fall within the limits of a single reactor fuel. The second artificiality is that the selected universe of fourteen reactors contains only light-water (both PWR and BWR) reactor fuel, and thus all of the reactor feedstock is the same type of low enrichment fuel. The third artificiality is that only a select set of radiochemistry data is included within SFCOMPO, and more specifically, those radiochemistry data that are most relevant to burnup credit methodologies rather than nuclear forensics. There may be other isotopic measurements that would be better discriminators or identifiers for spent fuel in the forensics domain.

SFCOMPO data sets had been used in a previous effort to explore the use of statistical methods to reveal patterns and associations in SNF data that had been repurposed for nuclear forensic applications. The success of this effort demonstrated that this modified version of SFCOMPO could be used as the foundation for a tabletop exercise that focused on class association involving an "unknown" SNF sample with finite, known families of isotopics. There are two specific modifications to the SFCOMPO data sets that enabled their use in *Galaxy Serpent*. First, uncertainty values for the data points needed to be determined. Since SFCOMPO data did not include measurement uncertainties, it was necessary to generate a robust set of uncertainty values. The uncertainties were a prerequisite both to generating the set of problems, namely data sets for each of the hypothetical seizures, and to performing any forensic evaluations of the problems. Determining and assigning uncertainty values to the PIE measurements in SFCOMPO proved to be a significant task and involved including uncertainties from published references associated with SFCOMPO data or the use of "best judgment" based on traditional analytical methods for the determination of uncertainties in similar fuel matrices.[8]

Secondly, based on the premise that the spent nuclear fuel isotopic compositions in the SFCOMPO database represent the entire universe of SNF for *Galaxy Serpent*, five forensics problems had been created based on actual SFCOMPO data. SFCOMPO data for a particular fuel pin measured at various positions (and burnup) were used to model the variation of the isotopic compositions as a function of fuel burnup. This mathematical model was then used to derive (i.e., to interpolate or extrapolate) isotopic compositions at other positions or different burnup values — simulating measurements obtained on samples at different times in the irradiation history of the fuel pin. Finally, a random adjustment was applied to these values representative of measurement noise, and these adjustments were consistent with the measurement uncertainties for the corresponding SFCOMPO data.[8]

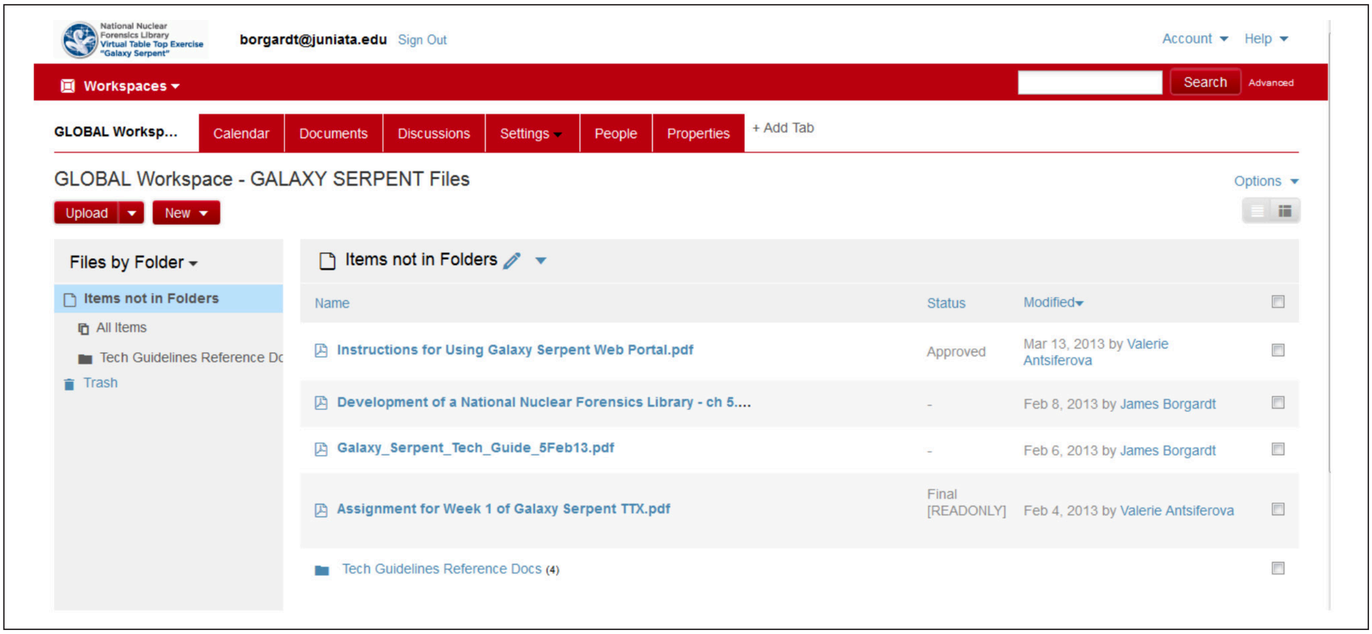
For *Galaxy Serpent*, SFCOMPO data was adapted for a nuclear forensic application, which involved families of isotopic correlations for specified reactors. As a result, it is important to realize there is no need to average any of the PIE values from samples pertaining to the reactors. For this forensic application, the PIE data from the samples are typically treated as discrete samples from a "smeared" reactor core "entity" for each of the fourteen reactors. Therefore, the geometric position data and information included in SFCOMPO for each sample is not relevant when creating the isotopic correlations that may distinguish among reactors or reactor classes. The correlations assume that the samples are representative of the isotopic compositions contained in a "smeared" reactor core as a function of exposure (i.e., neutron fluence).

These considerations and modifications led to a subset of fields from each original SFCOMPO data set, with the exclusion of identifying and positional data for the reasons noted above, and the addition of three fields associated with the assigned uncertainty, comprising the modified reactor data sets provided to each team. Specifically, each team was given data sets containing, for each data point, the following fields: the sample group, the isotope or isotopic ratio being measured, its actual value and units, the cooling time, and the ascribed uncertainty expressed as both 1-sigma and percentage. Actual reactor names in the SFCOMPO database were masked, renaming each after a moon of Saturn.

Exercise Play

The *Galaxy Serpent* international virtual tabletop exercise was designed to enable teams to use public domain data, have am-

Figure 1. Galaxy Serpent web portal workspace example



GLOBAL Workspace - GALAXY SERPENT Files

Items not in Folders

Name	Status	Modified	
Instructions for Using Galaxy Serpent Web Portal.pdf	Approved	Mar 13, 2013 by Valerie Antsiferova	☐
Development of a National Nuclear Forensics Library - ch 5....	-	Feb 8, 2013 by James Borgardt	☐
Galaxy_Serpent_Tech_Guide_5Feb13.pdf	-	Feb 6, 2013 by James Borgardt	☐
Assignment for Week 1 of Galaxy Serpent TTX.pdf	Final [READONLY]	Feb 4, 2013 by Valerie Antsiferova	☐
Tech Guidelines Reference Docs (4)			☐

ple time to work on Phase 1 and 2 tasks, and reach out to appropriate expertise as needed. To enable these factors during exercise play, a web-based approach was used because it provides easy accessibility for all teams, does not involve travel or material transport costs, does not involve analytical measurements, and enables teams to engage experts that may not usually be involved in ITWG activities. An in-person tabletop exercise format would not have been practical for *Galaxy Serpent*, because the Phase 1 and 2 tasks were usually not completed within hours or days, nor would the teams have been able to incorporate relevant expertise as needs arose during the exercise. Use of provided, published, public domain SNF data and information eliminated any sensitivities regarding country teams using their own materials data. As a result, teams only used their expertise during the exercise and were not required to use their own countries' data. It is recognized that when developing an NNFL, much of the materials information exists and may have likely been collected for other purposes. By using provided data in *Galaxy Serpent*, teams directly experienced organizing a small, model NNFL from existing data and information. Additionally, constraining the "universe" of nuclear reactors to those in the SFCOMPO database helped bound the problem so that the teams would formulate results in a finite amount of time. The exercise microcosm provided a model environment where ideas, concepts, and frameworks pertaining to NNFLs could be discussed and tested, allowing teams to effectively consider the process of creating an NNFL, while also applying it to a hypothetical seizure to see the potential value of an NNFL as an investigative tool. At the conclusion of the exercise, practical ideas and lessons from this *Galaxy Serpent* microcosm could be scaled up to address issues of creating or managing actual NNFLs.

Teams were provided with reference materials on nuclear forensics and NNFLs, including guidance documents drafted by the IAEA and the NNFL Terms of Reference from ITWG,[9] but not instructions on how to construct a NNFL. Each of the three to four teams participating in a given round was provided, through the dedicated web portal, data sets for three different reactors. This required the use of adjusted data sets for twelve of the fourteen possible SFCOMPO reactors. The reactors assigned to teams were scrambled for each round. The sole requirement was that each team be given a combination of PWR and BWR reactors. As noted, the team and reactor identities were masked; teams were named after galaxies and the reactors named after moons of Saturn (Table 2). The web portal was designed to have two workspace levels: one public and accessible to participants and observers, and one private and accessible only by members of a given team. This was arranged so that teams, if desired, could communicate anonymously through a public discussion forum (shown in Figure 1) to exchange challenges encountered, methodologies, access reference material, and the like. The private forum served to

Table 2a. Summary of SFCOMPO nuclear reactors and exercise pseudonyms (moons of Saturn)

	Reactor	Data Points	Reactor Type	Pseudonym
1	Calvert Cliffs No. 1	447	PWR	Anthe
2	Cooper	294	BWR	Atlas
3	Fukushima Daiichi-3	506	BWR	Enceladus
4	Fukushima Daini-2	1437	BWR	Daphnis
5	Genkai-1	123	PWR	Ijiraq
6	Gundremmingen	663	BWR	Hyperion
7	H.B. Robinson Unit 2	257	PWR	Iapetus
8	JPDR	1098	BWR	Janus
9	Mihama3	700	PWR	Mimas
10	Monticello	480	BWR	Pandora
11	Obrigheim	1035	PWR	Prometheus
12	Trino-Vercellese	1684	PWR	Siarnaq
13	Takahama-3	1227	PWR	Tethys
14	Tsuruga-1	270	BWR	Titan

Table 2b. Summary of Team and Seizure Pseudonyms, and Seizure Origins

Round	Galaxy Name	Team	Seizure	Origin of Seizure
1	Draco	Brazil	Clio	
1	Virgo	Australia		
1	Zwicky	Canada		Iapetus
2	Ursa	Japan	Erato	Siarnaq-1
2	Tucana	South Africa		
2	Sculptor	UK/AWE		
3	Andromeda	Sweden	Melpomene	
3	Keenan	Hungary		Daphnis
5	Centaurus	JRC/ITU	Terpsichore	Siarnaq-2

allow teams to discreetly communicate with exercise organizers, access provided data sets, and upload progress reports. The summary of the assignment of SFCOMPO reactors used in *Galaxy Serpent,* along with their aliases is shown in Table 2, and will be helpful in understanding the reactor names used in the articles which follow in this special issue. Table 2a links reactor pseudonyms with their identity in the SFCOMPO database, and gives the class and number of data points for each reactor. Table 2b provides the pseudonym for the seizure data set used in each round, and identifies the specific reactor of origin and the team which had this reactor as part of its model NNFL. For instance, in round 1 the Clio seizure originated from the Iapetus reactor which was assigned to the Zwicky galaxy (Canada). The two Siarnaq reactor seizures listed are distinct seizure data sets derived from the same reactor.

After being provided data sets, teams were given three to four weeks for Phase 1 in which to develop their model NNFL and were encouraged to share approaches or methodologies, as needed or desired. Some teams completed Phase 1 within one week, while others required additional time beyond the allocated time for a variety of reasons. In Phase 2, a hypothetical seizure was announced, and data for a hypothetical seizure, named after one of the Muses, provided. In a given round, all teams were provided identical seizure data, which originated from one of the nine to twelve reactors in play during that round. Each of the five created seizure data sets discussed earlier was used in one of the rounds. Teams used their model NNFL developed in Phase 1 to determine whether the seized material was or was not consistent with material in their model NNFL.

Teams were asked to determine whether provided data from a hypothetical seizure was or was not consistent with each of the three reactors in their model NNFL. When reporting their findings to exercise organizers, participants were instructed to use an ITWG established system of confidence levels. This defines evaluation criteria using a five-point convention for describing the similarity of samples: Conclusive Positive, Suggestive Positive, Inconclusive, Suggestive Negative, and Conclusive Negative. Determinations are made by evaluating how measurements and findings relate to legal statutes in relation to a defined "coverage parameter."[10] Teams use this scale in reporting confidence levels in their findings in the papers which follow.

Results

Teams successfully reported identification of the likely reactor from which the hypothetical seized SNF may have originated, as well as an evaluated set of "possibles." These evaluated problem solutions, obtained using conventional "isotope correlation techniques" (ICT), illustrate a clear, understandable, and defensible forensics capability for SNF. At least for this set of problem solutions, teams have demonstrated that the ability to identify the unknown materials from within the population of known samples is directly dependent upon the uncertainties in the data values and upon the gaps in the data values. Based on the assumptions made herein, in all cases, teams showed it was possible to downscale to a small number (to one, in some cases) of "possibles."[8]

More than 75 percent of the participant teams reported findings that were consistent with the origin of the hypothetical seizure. The remaining 25 percent did not report inconsistent results, but rather did not complete the exercise, for various reasons.

The exercise has been successful in a number of areas. Developing a NNFL containing nuclear data potentially has both national security and proprietary commercial sensitivities. The use of published, public domain data removes many of these concerns. *Galaxy Serpent* has also expanded the pool of experts aware of the use and potential efficacy of NNFLs, including reactor engineers, fuel experts, and statisticians. The web-based approach allowed a cost-effective method to advance the goals of the exercise, and also provided ITWG members with more opportunities to interact throughout a year, rather than limiting contact to ITWG annual meetings or reviewing of ITWG draft documents. A number of teams pursued parallel paths, such as statistical methods and isotopic correlation techniques, which yielded corroborating results.

While teams may have exhibited various levels of expertise and detail in working through the exercise, they were able to obtain useful and probative findings. Similar conclusions apply to the complexity of the developed model libraries; increased sophistication often facilitated greater resolution in assessing whether the seizure was or was not consistent with reactors in the model NNFL. However, it is critical to note that even a basic library proved valuable in providing critical insights as to the origin of the seizure.

The advantages and disadvantages, discussed earlier, associated with the repurposed SFCOMPO data did impact participants ascribing confidence levels to their findings. In an actual event, the analytical and investigate work would not occur dissociated from other communities, such as first responders, law enforcement, legal representatives, and policymakers. While the constrained universe, comprised of only low-enriched uranium (LEU) reactors, may have limited the range of sources teams had to consider, many note in their articles that the limited (in number of reactors, and samples within a reactor) and incomplete (in the variety of provided parameters) data sets presented challenges in assigning a confidence level. Nevertheless, despite these artificialities, all teams noted that the ability to compare data from a hypothetical seizure with a pre-established NNFL was essential in reaching conclusions.

Conclusions

The virtual, web-based *Galaxy Serpent* tabletop exercise demonstrated the efficacy of NNFLs in drawing inferences about the origins of a hypothetical seizure of spent nuclear fuel. It also showed that, however useful NNFLs proved, they would be even more effective when used in conjunction with an investigative effort involving many communities within a nation. A number of teams reached out to expertise outside their discipline for assistance, or in order to independently pursue multiple technical approaches. The collaborative option built into the exercise, via communication in the global workspace of the web-portal, was used by some teams to exchange methodologies and questions regarding interpretation of data sets, but was, in general, an underutilized facet of the exercise. This seemed to largely be due to the weeks-long timeframe of each phase allowing teams to progress at different paces, but may have also involved inherent sensitivities over the nature of the exercise. Several participants also preferred to communicate directly with exercise organizers to have such questions answered.

The universe of data sets were intentionally constrained to LEU reactors, which helped bound the problem for this exercise. The basic model NNFLs, composed of just three reactors, as well as their limited data sets, represented additional artificialities. Nevertheless, this exercise experience provides practical lessons as to the utility NNFLs can have, and how nuclear forensics may provide a powerful probative tool to help rule in or rule out data or information relevant to an investigation. Comparisons of the data from the hypothetical seizure with NNFLs helped each team quickly, in relation to a full-fledged inquiry, determine if the hypothetical seized SNF is or is not consistent with their holdings. Several teams also demonstrated that independently applied analytical methodologies confirmed findings. In an actual nuclear security event, the question "Is it ours?" may likely be one of the first questions asked by senior officials, and in the context of this exercise NNFLs proved to have high efficacy in addressing this key concern.

The exercise was successful in expanding the community of experts aware of nuclear forensics and NNFLs. The web-based format also allowed an international collaboration of scientists representing, all told, more than twenty countries. Participants found the exercise beneficial, instructive and insightful, and many requested a follow-on "*Galaxy Serpent 2.0*" exercise based upon a different class of nuclear material. Despite noted artificialities, the exercise proved valuable in engaging and expanding the existing nuclear forensics community of experts, and advanced the concept of national nuclear forensics libraries.

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Australia's Experiences in the Galaxy Serpent Virtual Tabletop Exercise

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Abstract

Australian Nuclear Science and Technology Organization (ANSTO) Nuclear Security Research Program staff participated in the International Technical Working Group on Nuclear Forensics (ITWG) Galaxy Serpent National Nuclear Forensics Library (NNFL) Virtual Tabletop Exercise. A model NNFL was established using spent fuel composition data from three reactors code-named Anthea-PWR (pressurised water reactor), Atlas-BWR (boiling water reactor) and Enceladus-BWR. The NNFL was compared with an intercepted sample (Clio) using statistical analysis from basic Microsoft Excel chart tools to the more complex multivariate analysis. Our results indicated that the Clio material was a suggestive negative match to have originated from our Galaxy. The exercise provided valuable lessons in the process of establishing an NNFL. These are fully discussed in the paper.

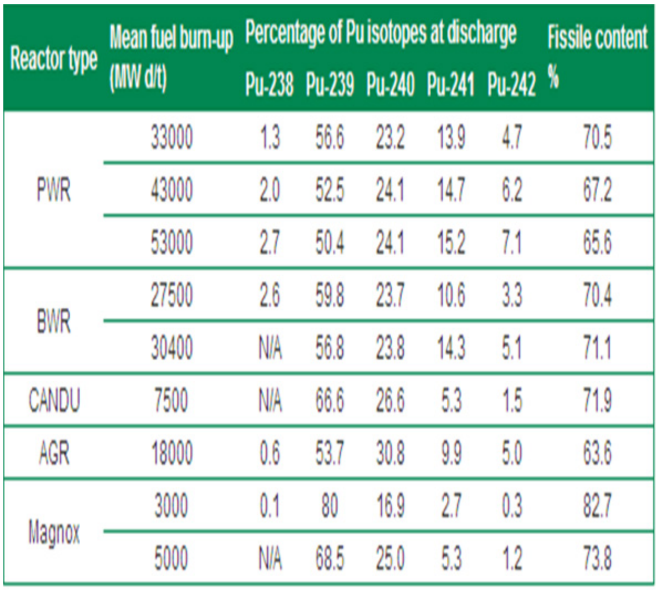
Introduction

Background

The Australian Nuclear Science and Technology Organization (ANSTO) houses a large proportion of Australia's Nuclear Forensics (NF) expertise and capabilities.[1] Consequently ANSTO, on Australia's behalf, participated in the International Technical Working Group on Nuclear Forensics (ITWG) Galaxy Serpent Virtual Tabletop Exercise (TTX).

The purpose of the TTX was to identify the strategies and technologies involved in establishing a national nuclear forensics library (NNFL). Different groups of countries participated in three separate rounds of the TTX. Australia participated in the first round of the TTX. Each round was composed of two phases: i) compilation of a model NNFL using three sets of spent nuclear fuel (SNF) isotopic data provided by the exercise organizers, and ii) comparison of data from a hypothetical seizure of SNF with the NNFL. The objective of the second phase was to determine if the seized material came from one of the three reactors in the model NNFL. This TTX was a blind test, i.e., participants were not told whether or not the unknown was comparable with their NNFL samples until after the completion of the TTX round. Further information of the TTX's overall objectives and structure can be found in the TTX Technical Guide for Players.[2]

Table 1. Example of plutonium composition produced from different sources[5]



Reactor type	Mean fuel burn-up (MW d/t)	Percentage of Pu isotopes at discharge					Fissile content %
		Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	
PWR	33000	1.3	56.6	23.2	13.9	4.7	70.5
	43000	2.0	52.5	24.1	14.7	6.2	67.2
	53000	2.7	50.4	24.1	15.2	7.1	65.6
BWR	27500	2.6	59.8	23.7	10.6	3.3	70.4
	30400	N/A	56.8	23.8	14.3	5.1	71.1
CANDU	7500	N/A	66.6	26.6	5.3	1.5	71.9
AGR	18000	0.6	53.7	30.8	9.9	5.0	63.6
Magnox	3000	0.1	80	16.9	2.7	0.3	82.7
	5000	N/A	68.5	25.0	5.3	1.2	73.8

Many factors can influence the isotopic composition of SNF. Pre-irradiation factors of significance include initial composition of fuel material, whether or not the fuel was derived from reprocessed material, impurities in the original ore, and days spent manufactured prior to irradiation.[3,4] During and post irradiation factors that affect isotopic composition include rate of fission in the fuel, irradiation time, position within a reactor core, cooling years and class of reactor (e.g., boiling water reactors [BWR] operate differently to pressurized water reactors [PWR]).[5]

Figure 1 shows a fuel assembly from a PWR, the most common reactor type. The position of each fuel pellet within a fuel assembly will receive unique thermal neutron fluence leading to a unique isotopic composition.[5] Table 1 shows that the amounts of particular isotopes generated during irradiation depend on: i) the type of reactor and ii) the level of burnup.[5] Of

Figure 1. Components of a rod cluster control assembly[5]

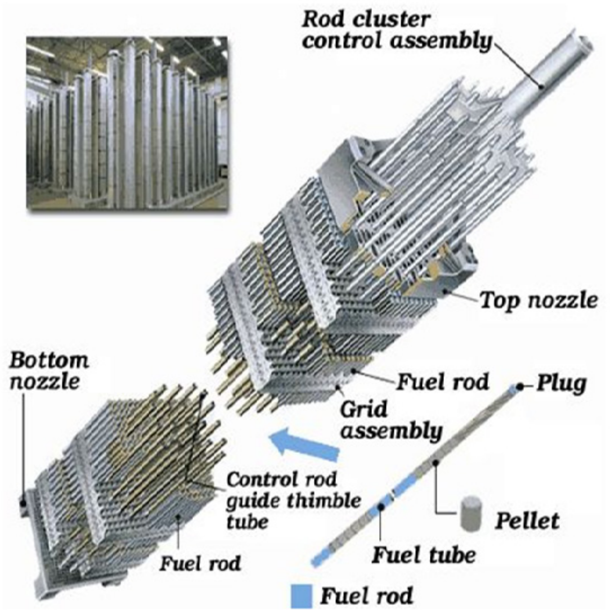
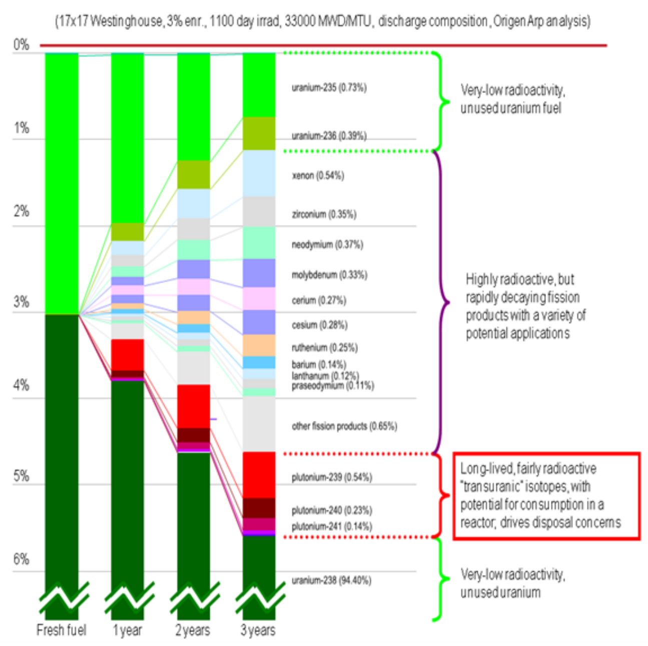


Figure 2. Example of isotopic composition of irradiated nuclear fuel in a conventional power reactor[6]



course, reactor operation can be optimized to produce power for changing demands.

Figure 2 demonstrates how significantly fuel composition changes over time of irradiation. Over irradiation time there is build-up of transuranic elements, decay chain products, and isotopes produced by nuclear fission.[6]

Regulations that apply in different countries can sometimes provide additional insight into where intercepted SNF may originate from. For example, some regulators have constraints on the amount of plutonium allowed to be present in spent nuclear fuel.

Isotopic composition of SNF is therefore a useful indicator of the provenance of SNF.

Method

Commencing the TTX, we were unsure which isotopes are important nuclear forensic markers for SNF, so we compiled information about the isotopes mentioned within the guidance notes[7] and within the data received (Appendix 1). Only certain isotopes of these were of relevance to this study. We have included the additional information in Appendix 1 as it may be useful for future NNFLs that contain materials other than SNF.

Interpretation of the Supplied Exercise Data

ANSTO was initially provided with isotopic data on irradiated fuel from three fictitious nuclear power reactors, code-named Anthea-PWR, Atlas-BWR, and Enceladus-BWR. The data was derived from information in the Spent Nuclear Fuel Isotopic Composition (SFCOMPO) database.[8] The exercise code name for this subset of Galaxy Serpent data was the Virgo Galaxy.

The exercise coordinators stated that the isotopic data from the reactors should be treated as *"discrete samples from a 'smeared' reactor core 'entity'"*.[7] We interpreted this to mean that the values we had been given were the average values of fuel pellets within a fuel rod, and assumed the fuel rod was positioned at unknown locations within the rod cluster control assembly (see Figure 1).

Later, isotopic data for a theoretically generated intercepted sample was provided and was to be treated as a *"nuclear fuel pin isotopically characterized at four points along the pin"*.[7] We interpreted this as four discrete pellets within a fuel rod. As outlined above, the positioning of fuel pellets within a rod, and the operational parameters the rod was exposed to (positioning, proximity to other rods at what periods of time, etc.) is important to know in an NNFL to be able to compare data to that from intercepted samples with greater accuracy.

Table 2. Example of the TTX supplied data from the Anthea-PWR set used to populate an NNFL

No	Data Type	Cooling Year	Values	1 sigma uncertainty	Unit	Uncertainty (% 1 sigma)
1	Am-241	6.7	0.377	0.018473	kg/MTU initial	4.90
2	Am-241	6.7	0.679	0.033271	kg/MTU initial	4.90
55	Pu-238/Total Pu(RateOfWeight)	6.7	0.0196	0.00044296	None	2.26
56	Pu-238/Total Pu(RateOfWeight)	6.7	0.0326	0.00073676	None	2.26
440	burnup(by Nd-148 method)	6.7	46.5	1.1625	GWD/MTU	2.50
441	burnup(by Nd-148 method)	6.7	37.3	0.9325	GWD/MTU	2.50

Isotopic data could be broken down into discrete "data types," dependent upon i) amount of an isotope in kg/MTU; ii) burnup of an isotope in GWD/MTU; iii) ratio of an isotope to either an isotope or element by weight; iv) a ratio of an isotope to either an isotope or element by atom precent; v) "build-up" of an isotope (^{236}U only); and vi) "depletion" of an isotope (^{235}U and ^{238}U only). Not all data types were available in all three data sets. Consequently we calculated as many variables as we could from the supplied data without making any assumptions about starting compositions and so on. Table 2 shows example components of a data set received in the exercise.

For each data type, two additional parameters were given: i) uncertainties (1 sigma) and ii) cooling years. In the third data set, the cooling years were all 0, suggesting the data set was modeled.

The organizers stated that the data provided was derived from analytical data, with specific information identifying the sample location in the reactor removed, thus treating the reactor as being homogenous or smeared for the purposes of the exercise. Many of the uncertainties provided with the data were uniform between data types, irrespective of the magnitude of the value; i.e., errors for some trace isotopes were the same as isotopes in much greater abundance when it would be expected that the error for trace isotopes would be much greater in value. Furthermore, the original SFCOMPO does not list associated errors with measurements. Consequently as we did not know how the uncertainties were calculated and questioned their integrity, we decided not to include the uncertainties when analyzing the data.

The "data types" and associated units received in the exercise are different from the data proposed to be used in the IAEA document Development of a Nuclear Forensic Library (in draft).[9] It was apparent the language used in the SFCOMPO database is designed for nuclear engineers, not forensic chemists. For example, the data provided expressed weights of isotopes as kg/MTU, MTU could be interpreted as metric ton of uranium, metric ton unit, or metric ton of heavy metal whereas these values would be more clearly expressed in SI units as g/g. The data types with "rate of weight" quoted after the isotopes in fact implied 'ratio of atoms." In addition to our forensic chemists, ANSTO nuclear engineers were not directly familiar with some of the various units.

An intercepted sample would be analysed by chemists. How then would the values be easily compared to the data entered by a nuclear engineer? When setting up an NNFL, the framework needs to be created in a way that when data is entered by a nuclear engineer, it would be converted for a forensic chemists' application, suggestively in the format found in Reference 9.

Creation of an NNFL

The isotopic data from the three reactors was compiled into an NNFL, which consisted of two Microsoft (MS) Excel worksheets. The first worksheet separated all of the data types and expressed units (provided and calculated) by columns, and samples belonging to a particular reactor separated by rows. The second NNFL library worksheet contained small subset of the first sheet composed of the eighteen isotopic correlations that are proven to be useful to mathematically articulate the differences between reactors or classes of reactors.[7] Table 3 shows a small subset of this worksheet. The worksheets were created in the best format to extract information readily into user generated MATLAB algorithms — our platform to conduct multivariate analysis (MVA). In an official NNFL, this information would be held as part of a relational database with other properties of SNF opposed to MS Excel files.

MS Excel Graphical Analysis of the NNFL Data

Prior to receiving information on the intercepted sample, we constructed 2D plots of the various data types from the three

Table 3. Isotopic correlations available from Virgo Galaxy reactors

Reactor	Sample number	U235% / U-236%	(Np-237/ U236)/ U-235%	(Pu/U238)/U235%	U235%/(Pu/U)
Anthe-PWR	1	1.347692308	0.200913242	0.019732891	51.10851809
	2	0.463768116	0.731884058	0.065831557	15.23809524
	3	0.89244186	0.331414287	0.030667225	32.8342246
Atlas-BWR	1	1.511221945	0.160901377	0.013940148	72.48803828
	2	4.530201342	0.043251305	0.004783467	212.5984252
	3	1.333333333	0.168717921	0.014759553	68.4144819
Unknown Clio-1	1	3.042896608	0.056679288	0.00484989	209.3833343
Unknown Clio-2	2	1.250392855	0.156975926	0.012935539	78.12343555
Unknown Clio-3	3	0.904797384	0.225616535	0.01880911	53.64692536
Unknown Clio-4	4	1.802190937	0.102894731	0.008253987	122.6554573

reactors. This enabled us to see correlations between data types and to see which data types showed the greatest variations between reactors.

Linear regression of ^{236}U (wt %) versus ^{235}U (wt %) of the three reactors were used to ascertain the starting enrichment of ^{235}U in their fuel, or to determine whether the fuel was of reprocessed nature.

Multivariate Statistical Analysis Techniques

Two multivariate statistical analysis techniques were used to evaluate the data from the reactors. Firstly, Principal Component Analysis (PCA) was used to assess the reactor data. PCA has been widely used for evaluating relationships in the underlying data and for dimensionality reduction purposes.[10] PCA generates Principal Components (PCs), which are a new set of variables that are uncorrelated and are arranged so that the first few retain most of the variation in all of the original variables (data types). In this work, PCA was used to both evaluate the relationships of the underlying reactor data and to reduce the dimensionality of the problem. The PCA method used the first three PCs to evaluate whether the data would separate into their respective reactors. A 3D scatter plot of these PCs (PC1 vs. PC2 vs. PC3) was used to visualize the distribution of the different reactor data. While the PCs generated by PCA may explain a large proportion of the variation in the data, they may not represent the optimized separation between different reactors. For a more detailed description of PCA, a comprehensive review has been performed by Jolliffe.[10]

The benefit of another multivariate analysis technique, Fisher Linear Discriminant Analysis (FLDA), is that it can determine the optimized loading coefficients (LCs) that maximizes the separation between two known groups of data. FLDA can be used for dimensionality reduction purposes but is also typically used as a classification technique. Previous use as a classification technique has covered a range of problems including: gamma-ray identification,[11,12] face recognition,[13] and medical image analysis.[14] In this work a data group represents the data from one particular reactor, so FLDA can determine an optimized separation between the reactors. The generated LCs are effectively a set of weighting factors for each original data type. The projection of the original data types by the LCs creates a single data point for the associated reactor. In this work, LC1 corresponds to the data projected by the Anthea-PWR LCs, LC2 corresponds to the data projected by the Atlas-BWR reactor LCs and LC3 corresponds to the data projected by the Encleadus-BWR LCs. A more detailed mathematical explanation of FLDA can be found in Reference 11.

In this study, the data groups were a subset of the initial sample data from the three reactors. In order to increase the discriminatory power of PCA and FLDA, the data was standardised (i.e., divided each data type by its standard deviation), which gives a variance within each data type of one, ensuring that prominent isotopes such as ^{238}U and ^{235}U do not dominate the analysis.

Analysis Strategies

Analysis strategies were initially based on the guidance provided within the NNFL TTX Technical Guide for Players.[7] The guide had provided eighteen isotopic correlations based on surrogates for fluence, proven to be useful in separating SNF data. '^{235}U% vs ^{240}Pu/^{239}Pu' is one example of the eighteen provided correlations. Where data was available, each 'isotopic correlation' was initially plotted directly in 2D scatter plots using MS Excel. When applying MVA, each broken down component of these correlations was used as either a new or existing data type. For example in the above correlation ^{235}U% was one data type and ^{240}Pu/^{239}Pu as another. We initially plotted all of the data types consistent between the three reactors. Separately we plotted all of the data types belonging to components of the eighteen isotopic correlations. Different analysis strategies using MVA of only select data types are described below. Conclusions were made using the jargon from the ITWG Guideline for Graded Nuclear Forensics Decision Framework (in draft).[15]

Figure 3. A 2D Scatter plot of the isotopic correlation ^{241}Pu/^{240}Pu vs ^{240}Pu/^{239}Pu comparing variance of "Vigro Galaxy's" three reactors

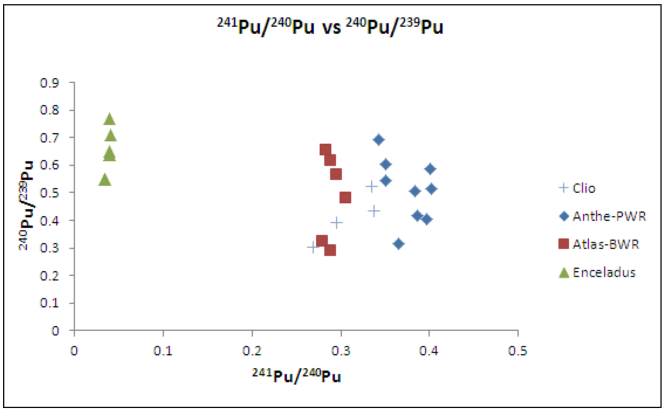


Figure 4. A 2D scatter plot of ^{235}U% and ^{236}U%, using linear regression to determine initial ^{235}U%

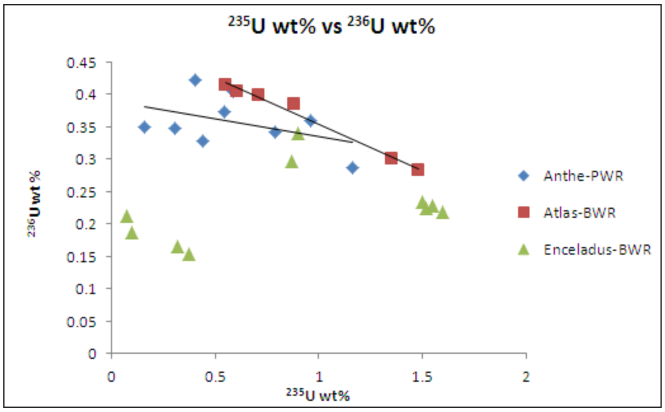


Figure 5. A bar plot generated by PCA of the ^{235}U%, ^{236}U%, ^{240}Pu/^{239}Pu, 241Pu/^{240}Pu, ^{242}Pu/^{240}Pu, and ^{242}Pu/^{241}Pu data types between the three reactors

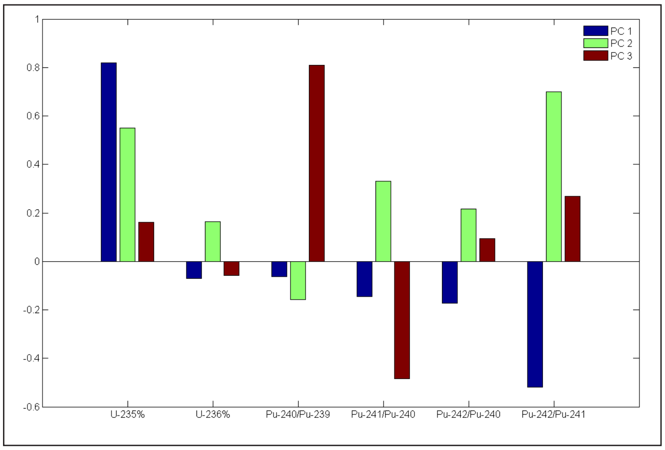
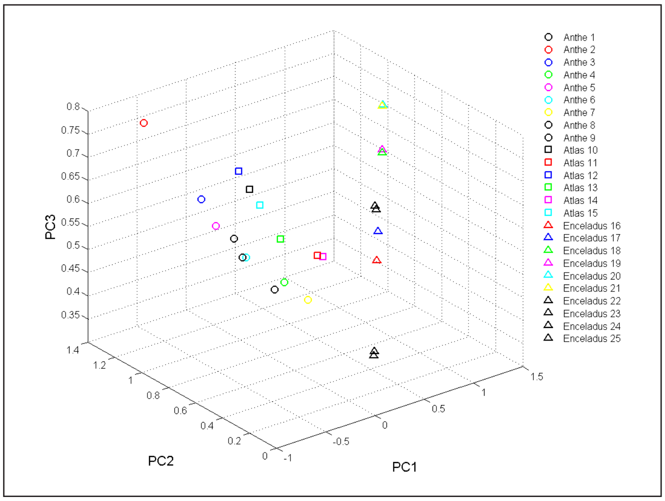


Figure 6. A 3D scatter plot using PCA of the ^{235}U%, ^{236}U%, ^{240}Pu/^{239}Pu, ^{241}Pu/^{240}Pu, ^{242}Pu/^{240}Pu, and ^{242}Pu/^{241}Pu data types between the three reactors



Results

MS Excel Analysis of the NNFL Data

In most cases the 2D plots looked like unstructured scatter diagrams; however, in some cases the data for the three reactors showed distinct separation. For example, Figure 3 shows a plot of the (^{241}Pu/^{242}Pu) versus (^{240}Pu/^{239}Pu) ratios in which the data for the three reactors is clearly separated. In other cases these scatter plots highlighted whether the SNF pre-irradiation were of a reprocessed origin. For example Figure 4 shows a plot of ^{235}U% versus ^{236}U%, which includes lines of best fit (derived via linear regression of the data). In conventional power reactors the enrichment of unirradiated fuel is typically between 3 percent and 5 percent ^{235}U and the ^{236}U content is not present (unless the fuel is derived from reprocessing in which it is at very low concentrations).

The lines of best fit for the Anthea-PWR and Enceladus-BWR in Figure 4 do not extrapolate into the 3 percent and 5 percent ^{235}U range. This suggests that the Anthea-PWR and Enceladus-BWR pre-irradiated fuel is recycled or reprocessed.

Application of PCA to the NNFL Data

PCA was successfully applied to our NNFL data sets and allowed us to identify criteria that clearly differentiate between the three reactor data sets. We included all data types excluding 'ratios of atoms' where weights are provided. We plotted various combinations of data types using PCA. After many iterations, we found that the following data types showed the most separation between the three reactors: ^{235}U%, ^{236}U%, ^{240}Pu/^{239}Pu, ^{241}Pu/^{240}Pu, ^{242}Pu/^{240}Pu, and ^{242}Pu/^{241}Pu.

Figure 7. A bar plot generated by FLDA of the ^{235}U%, ^{236}U%, ^{240}Pu/^{239}Pu, ^{241}Pu/^{240}Pu, ^{242}Pu/^{240}Pu and ^{242}Pu/^{241}Pu data types between the three reactors and intercepted sample Clio

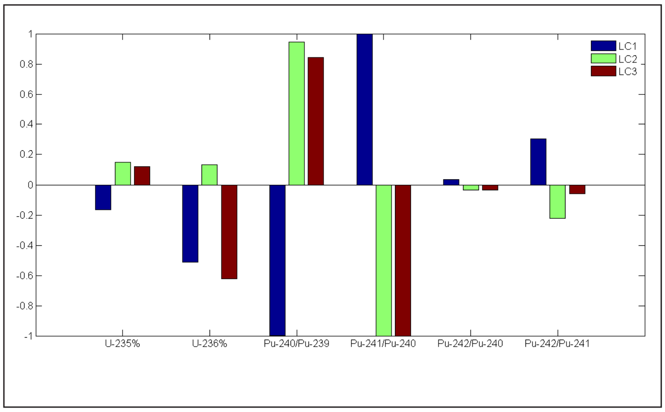


Figure 8. A 3D scatter plot using FLDA of the ^{235}U%, ^{236}U%, ^{240}Pu/^{239}Pu, ^{241}Pu/^{240}Pu, ^{242}Pu/^{240}Pu and ^{242}Pu/^{241}Pu data types between the three reactors and intercepted sample Clio

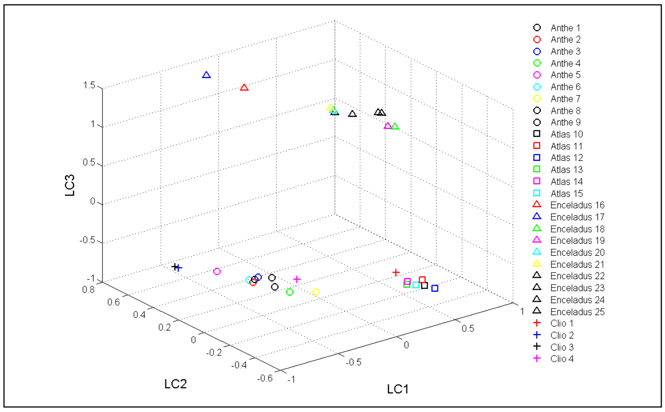


Figure 9. A 2D scatter plot of the isotopic correlation ^{241}Pu/^{240}Pu vs ^{240}Pu/^{239}Pu comparing variance of "Vigro Galaxy's" three reactors with intercepted sample Clio

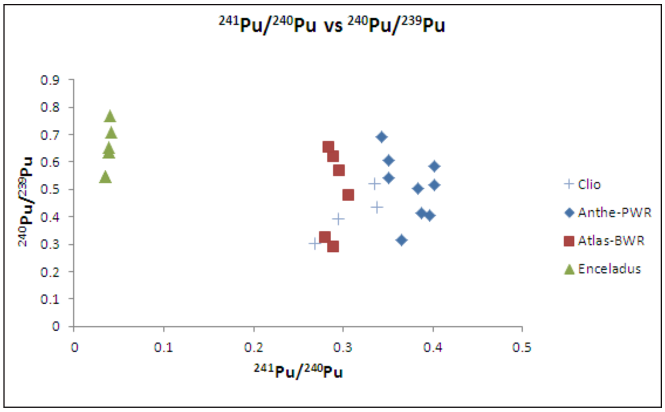


Figure 5 shows a bar plot depicting weighing factors of the first three PCs for the six isotopic correlation data types. These weighing factors show the contribution from each data type on each PC. For example, data from the ^{240}Pu/^{239}Pu and ^{241}Pu/^{240}Pu ratios dominated separation on the PC3 axis (due to their higher weighing factors).

Figure 6 shows an example of a 3D scatter plot. It contains three distinct groupings that do not intersect each other (circles, squares and triangles), each shape representing samples from each of the three reactors.

Comparison of the Intercepted Sample with the NNFL Data Using FLDA and MS Excel

The intercepted sample data Clio was received and entered into the NNFL MS Excel worksheets. Where the raw data was available, missing data types were calculated.

Some of the data types received in the Clio sample did not match data types associated with the three reactors in our galaxy. Although we entered these into our NNFL, this data was not used in the MVA techniques described below as we could not compare like with like data. In a real scenario, the data types not available for both an intercepted sample and a potential source reactor would still prove useful to nuclear engineers assisting nuclear forensic experts as they could use this data to help model theoretical values of isotopic compositions in pre and post-irradiated fuel using tools such as the SCALE (Standardized Computer Analyses for Licensing Evaluation) code system.[16]

Figure 7 shows a bar plot depicting weighing factors of the FLDA LCs for the six isotopic correlation data types used in Figure 8. It was observed that the ^{239}Pu/^{240}Pu and ^{241}Pu/^{240}Pu ratio weighing factors contributed to the separation the most on LC1, LC2 and LC3. The Clio samples were included into the 2D MS Excel plot for these ratios (Figure 3); however, the Clio sample showed no distinct separation from either the Anthea-PWR or Atlas-BWR (Figure 9).

Figure 8 shows a 3D scatter plot using FLDA. As with PCA, each shape (circles, squares, and triangles) represents samples from each of the three reactors. The Clio samples are depicted with plus symbols. The isotopic correlation data types used were i) available between all reactors and the unknown sample; and ii) yielded the best clustering of the three reactors prior to including the unknown sample in the MVA analysis (Figures 5 and 6). Figure 8 shows clear separation of the Enceladus-BWR from the Clio sample and other two reactors on the LC3 axes. The Clio samples were unevenly distributed around both the Anthea-PWR and Atlas-BWR clusters, at this

Figure 10. A 2D scatter plot using FLDA of the 'isotope by weight' data types between the Anthe and Atlas reactors, and seized sample

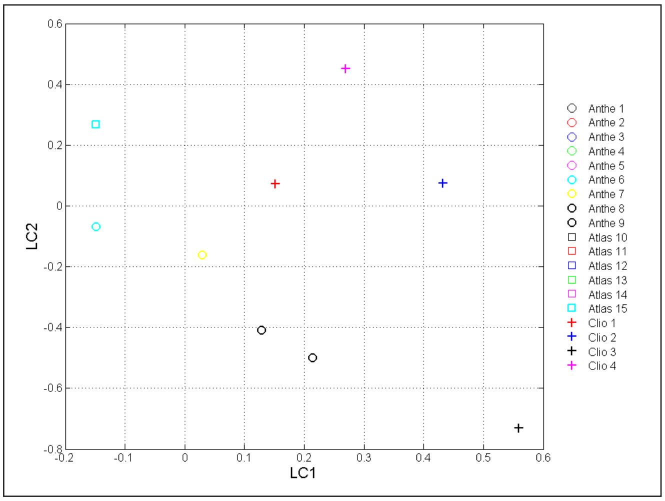
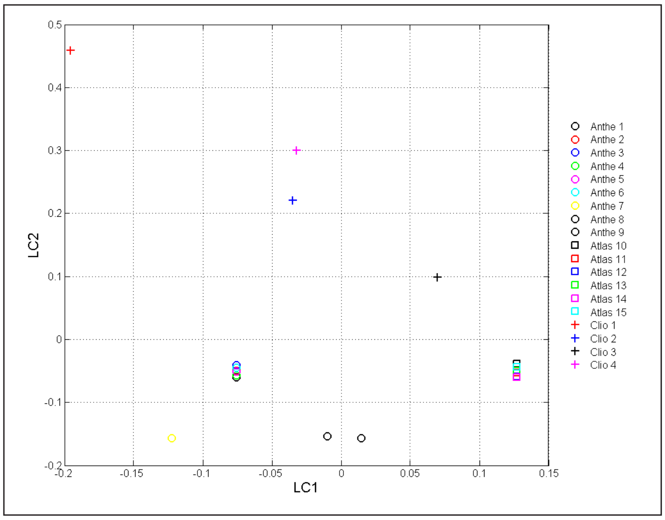


Figure 11. A 2D scatter plot using FLDA of the plutonium ratios, total Pu/^{238}U, total Pu/total U, 99Tc/ U, ^{241}Am/total U, ^{237}Np/total U, ^{237}Np/ ^{236}U and ^{236}U/^{238}U data types between the Anthe and Atlas reactors, and seized sample



stage more significantly present around Anthea-PWR based on separation contributed from the LC1 and LC2 axes.

These results allowed us to exclude the Enceladus-BWR from future FLDA plots. This was useful because, as outlined earlier, many of the data types available for Anthea-PWR, Atlas-BWR, and the unknown samples Clio were not available for Enceladus-BWR. In the following results, an iterative process was followed using the data types and isotopic correlations available for just Anthea-PWR, Atlas-BWR and the intercepted Clio samples.

Figure 10 shows an FLDA analysis where the entire set of exercise supplied isotope 'by weight' data types are included. On the LC1 axes, the unknown Clio samples overlap the Anthea-PWR; however, it was inconclusive as to whether the Clio samples were part of the cluster. The LC2 axes shows overlap of the unknown Clio samples to both reactors.

Attempting to further resolve clustering between the unknown samples and the reactors, only the most abundant transuranic isotopes in SNF were analyzed; however, this resulted in a greater overlap of the unknown sample between the reactor clusters (figure not shown).

A second analysis strategy involved assessing the following data types: i) amount of an isotope in kg/MTU; ii) burnup of isotope in GWD/MTU; and iii) ratio of isotope to an isotope. This also resulted in overlapping Clio samples to both reactors on both of the LC1 and LC2 axes (figure not shown). A third strategy yielded similar results for the FLDA analysis where all of the ratio data sets were analyzed (figure not shown).

From previous iterations, we had noted greater reactor clustering when using the Pu and Np ratios. Figure 11 shows the reanalyzed ratio data sets using only the plutonium ratios, total Pu/^{238}U, total Pu/total U, ^{99}Tc/U, ^{241}Am/total U, ^{237}Np/total U, ^{237}Np/^{236}U, and ^{236}U/^{238}U. On the LC2 axes the unknown sample appears separate to both the Anthea-PWR and Atlas-BWR. On the LC1 axes, it is inconclusive as to whether the intercepted sample belongs to the Anthea-PWR, and is a suggestive negative match to the Atlas-BWR.

Table 4 summarizes the results of our analyses, including the figures not included.

Discussion

Seized material was assessed using isotopic correlations against SNF from reactors within the Virgo Galaxy using MVA. We inferred a suggested negative match of seized material to our galaxy — most of the characteristics disagree, a few may agree.

The Enceladus-BWR resulted in a conclusive negative match to the provenance of the seized sample — all of the characteristics disagreed. All of the MVA strategies giving the best separation between the reactors resulted in no overlapping of the seized samples with the Enceladus-BWR sample clusters. A few MVA strategies with poor separation ability did show similar overlap with other reactors, suggesting these data variables are consistent between most PWRs and BWRs (figures not shown). These were typically strategies involving uranium isotope data types (masses and ratios).

Table 4. A summary of the data types used in each statistical analysis to determine whether the intercepted sample Clio matched a reactor from the Virgo Galaxy

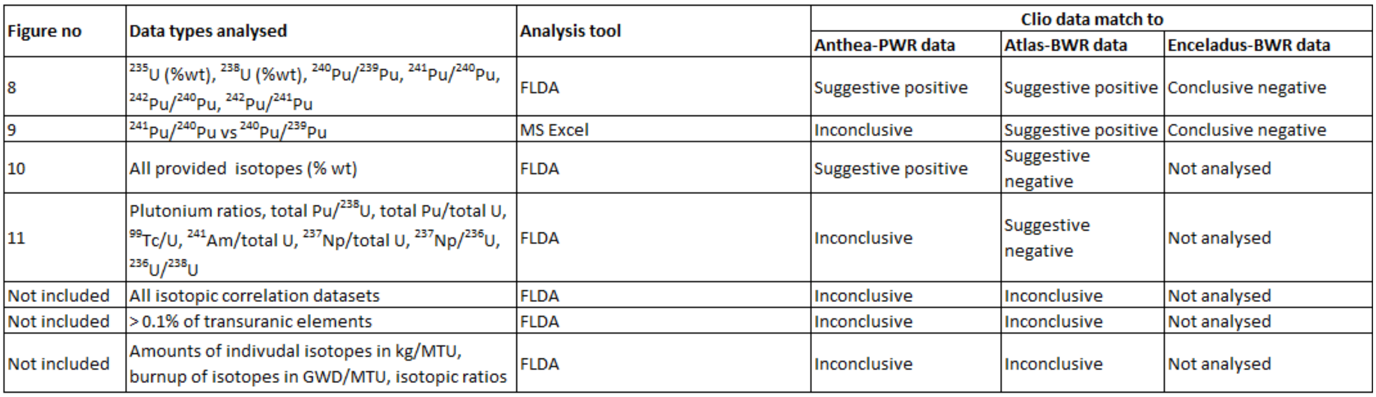


Figure no	Data types analysed	Analysis tool	Clio data match to		
			Anthea-PWR data	Atlas-BWR data	Enceladus-BWR data
8	^{235}U (%wt), ^{238}U (%wt), ^{240}Pu/^{239}Pu, ^{241}Pu/^{240}Pu, ^{242}Pu/^{240}Pu, ^{242}Pu/^{241}Pu	FLDA	Suggestive positive	Suggestive positive	Conclusive negative
9	^{241}Pu/^{240}Pu vs ^{240}Pu/^{239}Pu	MS Excel	Inconclusive	Suggestive positive	Conclusive negative
10	All provided isotopes (% wt)	FLDA	Suggestive positive	Suggestive negative	Not analysed
11	Plutonium ratios, total Pu/^{238}U, total Pu/total U, ^{99}Tc/U, ^{241}Am/total U, ^{237}Np/total U, ^{237}Np/^{236}U, ^{236}U/^{238}U	FLDA	Inconclusive	Suggestive negative	Not analysed
Not included	All isotopic correlation datasets	FLDA	Inconclusive	Inconclusive	Not analysed
Not included	> 0.1% of transuranic elements	FLDA	Inconclusive	Inconclusive	Not analysed
Not included	Amounts of indivudal isotopes in kg/MTU, burnup of isotopes in GWD/MTU, isotopic ratios	FLDA	Inconclusive	Inconclusive	Not analysed

The intercepted sample belonging to the Atlas-BWR resulted in a suggestive negative match — most of the characteristics disagree, a few may agree. Analysis strategies employed in Figures 8, 10, and 11 had shown limited or no overlap of the seized sample with Atlas-BWR sample data. Other analysis strategies (figures not shown) showed overlaps of seized sample with Anthea-PWR and Atlas-BWR samples. None of the FLDA iterations identified an overlap of seized sample isolated to just the Atlas-BWR. We therefore qualitatively concluded a suggestive negative match of the seized sample originating to the Atlas-BWR.

The Anthea-PWR resulted in an inconclusive match — multiple characteristics are in conflict regarding similarity and dissimilarity. Many of the FLDA analysis strategies resulted in the overlap of seized sample data with the Anthea-PWR (Figures 8, 10, and 11); however, in such scenarios, seized sample data is also distributed into other reactor clusters and outside reactor clusters entirely. The data for the reactors and seized sample were statically limiting to either exclude or include the provenance of a seized sample to a given reactor with a quantifiable level of confidence using FLDA.

Throughout our MVA iterations, we had learnt for our given reactor data set, the plutonium isotope ratios, particularly the ^{241}Pu/^{240}Pu, ^{242}Pu/^{240}Pu ratios yielded the most separation in their data sets (determined from biggest influence on weighing factors). This was followed by the ^{235}U%, ^{236}U%, and ^{237}Np/^{236}U. Although these data types may show the best separation for our given data set and application, it is important not to assume these data types are optimal for future MVA applications on SNF as: i) the values given have limited operational information associated with the values (other than cooling years); and ii) other data types not available to use in this exercise may yield an even better separation.

Although analyses were not statistically conclusive in proving or disproving the provenance of the seized sample from all reactors within the Virgo Galaxy, the analyses were able to exclude the seized sample from the Enceladus-BWR, and likely exclude the Atlas-BWR. In a real world scenario, such findings would complement other data such as physical characteristics (dimensions, densities) and other case related intelligence in order to forensically include or exclude suspect reactors to a seized fuel samples provenance.

It is important that forensic organizations plan to manage the expectations of their state authorities' pre- and post-incident with regard to realistic timelines in a real scenario. Under the scope of heavy regulatory and safety constraints, the time to i) isotopically characterize a seized sample; ii) request nuclear engineers to conduct theoretical isotope modelling of the seized sample; iii) add the information to a pre-generated NNFL; and iv) apply FLDA analysis strategies to help determine a seized samples provenance should all be well documented and agreed to between forensic organizations and state authorities.

State authorities seek evidence that provide probative value in a court of law. A comprehensive NNFL provides a database allowing nuclear forensic scientists to give statistical weighing to their comparative findings of a seized sample, as is currently done for DNA analysis. DNA evidence is a powerful tool for law enforcement as it provides a statistical measure of the probative significance of a match between suspect and sample. A random match probability can then be generated, which expresses the expected frequency of that DNA profile in a given population, therefore the chance of the match occurring coincidentally.[17]

Another important point is the sourcing of information used to populate an NNFL. In DNA databases there are set guidelines detailing best practices, which loci within the DNA to identify and quantitate. The database also includes standard definitions of what is a match versus no match. These too are things an NNFL should consider.

Nuclear forensic organizations need to have procedures in place to allow questions to be directed to subject matter experts. The NNFL concept provides an approach to facilitate such procedures. If the NNFL is not properly set up or supported by subject matter experts however, it can be a hindrance opposed to an asset.

Conclusions

- A basic NNFL can be readily generated using common software (such as MS Excel).
- If available, MVA techniques can provide additional insight to MS Excel analysis.
- Use of common units (SI) would aid communication and conversations between stakeholders (e.g., between different professional groups such as nuclear engineers and the forensics community; and between trusted partner countries).
- In this study we were advised that the data set for each sample at a given time was the average smeared set of values for an entire reactor core. We compared these reference data sets with data sets for discrete pellets. In a real scenario, the data in an NNFL would be data from discrete pellets but would include positional and operational information. Consequently it may be more difficult to make definitive findings in the real world.
- The experimental results from this study showed that the "unknown" was a suggested negative match to have come from any of the reactors included in the Virgo Galaxy NNFL.

Grant Griffiths received his BSc (Hons) at the Children's Medical Research Institute Westmead Australia and University of Western Sydney. Prior to his research career, Griffiths worked in a variety technical roles as a laboratory technician/ method developer at Lubrizol International, Orica Ground Water Treatment plant, and the Australian Nuclear Science and Technology Organization (ANSTO) in the Waste Operations division. In 2010 he became the Radiochemist Laboratory Manager for the Institute for Materials Engineering for multiple labs at ANSTO, including the Nuclear Forensics Research Facility in 2012. In addition to maintaining regulatory and safety requirements for staff working in the radiochemical laboratories he supervises, Griffiths has since co-authored publications over a broad range of research disciplines of life sciences and material sciences.

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David Hill is manager, nuclear security science at the Australian Nuclear Science and Technology Organisation (ANSTO). He is responsible for the promotion of ANSTO's capability in the fields of national security, nuclear forensics and related sciences, providing formal coordination and management of national security effort across and on behalf of ANSTO. His cur-

rent role advances previous work as ANSTO's National Security Research Program Leader focused on building world-class capabilities in radiation detection, environmental monitoring for nuclear safeguards, nuclear forensic research, and providing research support and services for the IAEA Australian emergency services, border security and law enforcement agencies. He is the current chair of the Nuclear Forensics Working Group for the Implementation and Assessment Group of the Global Initiative to Combat Nuclear Terrorism. Hill graduated from the University of NSW with a BSc and has more than thirty-five years' experience in analytical chemistry. He was initially employed by ESSO, where he worked in the Exploration Division Palaeontology and Geochemistry laboratories then moving to Petroleum Products in various quality assurance positions becoming Chief Chemist Australia in 1986. He joined ANSTO in 1988 as laboratory manager for the Environmental Chemical Analytical Laboratory and managed the ANSTO Institute of Environmental Research Inorganic, Organic, Radiochemistry and Tritium Analytical laboratories. In 2002 he was invited to join Standards Australia, and currently serves as chair of the spectroscopy committee.

Dr. Katherine L. Smith (Kath) is currently the Senior Advisor in International Relations in the Government International and External Relations division of the Australian Nuclear Science and Technology Organization (ANSTO). In this role she facilitates the relationships between ANSTO and its national and international stakeholders, assists in coordinating national security activities across ANSTO and contributes to research on ceramics and nuclear forensics. From February 2008 to July 2011, Smith was the counsellor for Nuclear Science and Technology (Counsellor, Nuclear) at the Australian Embassy in Washington, DC, USA. This role was fully funded by ANSTO and included both diplomatic/policy and research components. Smith's research interests were centered on the characterization, properties, and radiation response of nuclear waste forms and related materials. Previously Smith was a principal research scientist and group leader in the Institute of Materials Engineering at ANSTO, where she undertook research in radiation damage effects and had oversight of various research groups including those working in: nuclear forensics, radiation detectors and materials characterisation. Smith has a PhD in Physics and BSc (Hons) from Monash University.

APPENDIX 1

Appendix 1. Significance of particular isotopes

The data provided by the exercise organizers included some of these isotopes. Others are included as they might be useful when establishing a national nuclear forensics library.

Isotope Layout of characteristics listed within this table	**Significance** Fissile, Fissionable, Fertile, stable Natural abundance Created by… fission of… Unique characteristics/comments
^{235}U	Fissile Natural abundance 0.72 % Enrichment level depends on application. For example: Conventional enriched power reactor fuel contains 3-5% ^{235}U Low-enriched Uranium (LEU) < 20% ^{235}U Highly enriched Uranium (HEU) > 20% ^{235}U
^{234}U	Fertile Natural abundance 0.0055% Decay product of ^{238}U (alpha decay) Higher amounts of ^{234}U are found in enriched and depleted uranium compared to natural uranium due to enrichment processes that separate the lower uranium isotopes from ^{238}U
^{236}U	Weakly fertile Natural abundance: $< 1x10^{-10}$ % Generated by ^{235}U (n, γ) An effective burnup marker in nuclear forensic applications as it is not fissile and is weakly fertile. With supporting data, can be used to indicate initial ^{235}U enrichment in irradiated fuels.
^{238}U	Fertile and fissionable Natural abundance: 99.2745% Captures a neutron and becomes (indirectly) ^{239}Pu
^{239}U	Fissile material, short lived isotope Generated by ^{235}U (n,γ)
^{238}Pu	Fertile and fissionable Generated by ^{238}Np (beta decay), ^{239}Pu (n, 2n) and ^{242}Cm (alpha decay) Relatively low yield in power reactors compared to other Pu isotopes
^{239}Pu	Fissile Generated by ^{239}U beta decay to ^{239}Np, then beta decay to ^{239}Pu Characteristic marker of fluence received in a reactor
^{240}Pu	Fertile and fissionable Generated by ^{239}Pu (n,γ) Characteristic marker of fluence received in a reactor
^{241}Pu	Fissile Generated by ^{240}Pu (n,γ) Characteristic marker of fluence received in a reactor
^{242}Pu	Fertile and fissionable Generated by ^{242}Pu (n,γ) Characteristic marker of large amounts of fluence received in a reactor

^{241}Am	Fissile Generated by ^{241}Pu (beta decay), Under neutron irradiation, ^{241}Am readily captures neutrons and transforms to ^{242}Am. In a stable environment, the weight % of ^{241}Am builds up steadily over time, consequently it is a characteristic marker of how long material has been out from a fissile environment.
^{137}Cs	Fission product ~6.3% fission yield from ^{235}U May be used as a burnup indicator for fuel elements working at relatively low temperature ($<500^{o}$C). Short-term heat emissions caused by ^{137}Cs in spent fuel is a limiting factor for geological storage.
^{148}Nd	Fission product, stable isotope ^{148}Nd is a useful indicator of burn up as its fission yield is identical for ^{235}U and ^{239}Pu.
^{135}Cs	Fission product Under neutron irradiation, ^{135}Cs levels are low because its precursor ^{135}Xe is a good neutron absorber, which transmutes rapidly to stable ^{136}Xe. In a stable environment, ^{135}Cs levels increase steadily over time.
^{237}Np	Fissile Long-lived isotope, half-life of 2 million years considered as waste "bottle neck." Produced in the nuclear reactor from neutron irradiation of ^{235}U and ^{238}U. Can be used in nuclear fission reactions and has similar critical mass as ^{235}U. About 4-5% of ^{237}Np is found in spent nuclear fuel as Pu discharge.
^{239}Np	Short-lived isotope Produced from neutron capture of ^{238}U (n, γ)
^{79}Se	Fission product Can be used as a marker to determine ^{235}U burnup
^{126}Sn	Fission product Can be used as a marker to determine ^{235}U burnup
^{90}Sr	Fission product May be used as a burnup indicator for fuel elements working at relatively low temperature ($<500^{o}$C). Short-term heat emissions caused by ^{90}Sr in spent fuel is a limiting factor for geological storage. A huge amount produced during nuclear weapon testing. Used as a radioactive tracer in medical and agriculture studies.
^{99}Tc	Fission product Short-lived parent ^{99m}Tc is a widely use radioisotope in medical diagnostic applications.
^{95}Zr	Fission product Short-lived isotope found in material recently discharged from a reactor or after fission reaction.
^{95}Nb	Fission product Short-lived isotope found in material recently discharged from a reactor or after fission reaction.
$^{'x'}$I	Fission product
$^{'x'}$Xe	Fission product Natural abundance (of radioactive species): ^{136}Xe at 8.9%
$^{'x'}$Kr	Fission product Natural abundance (of radioactive species): ^{35}Kr at 0.35%

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The Use of a National Nuclear Forensic Library in Order to Identify Unknown Seized Nuclear Material

Brazil's Participation in the Galaxy Serpent Exercise

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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.

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

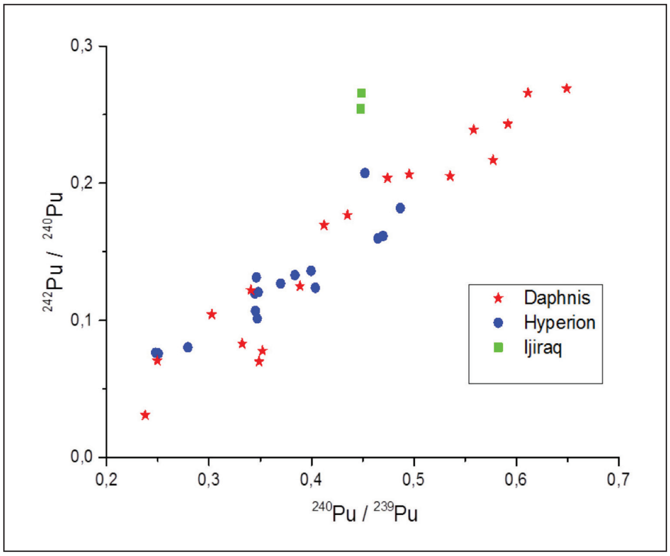


Figure 2. Isotopic Correlation: ^{238}Pu/ (^{239}Pu+^{240}Pu) versus ^{240}Pu/^{239}Pu

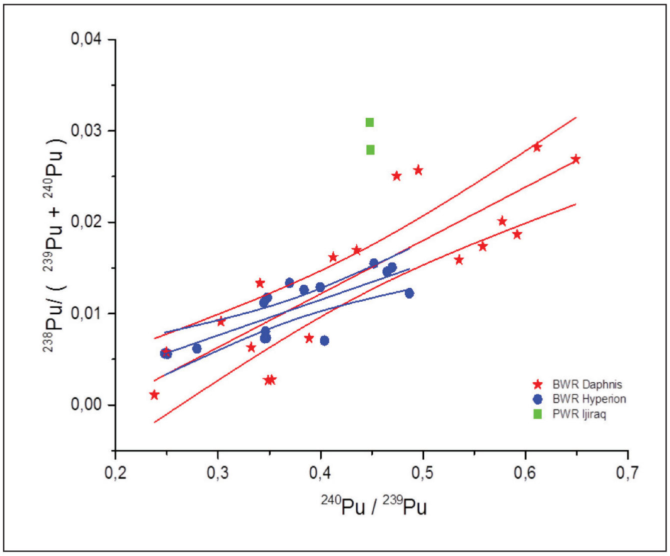
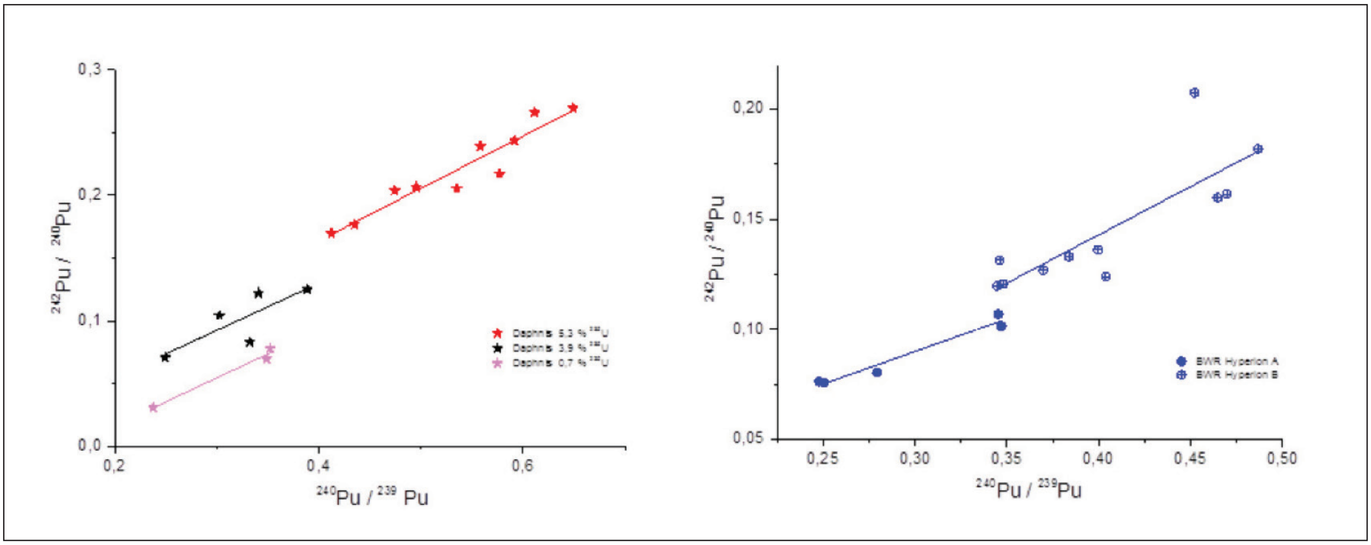


Figure 3. Daphnis and Hyperion: ^{242}Pu/^{240}Pu versus ^{240}Pu/^{239}Pu



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).

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.

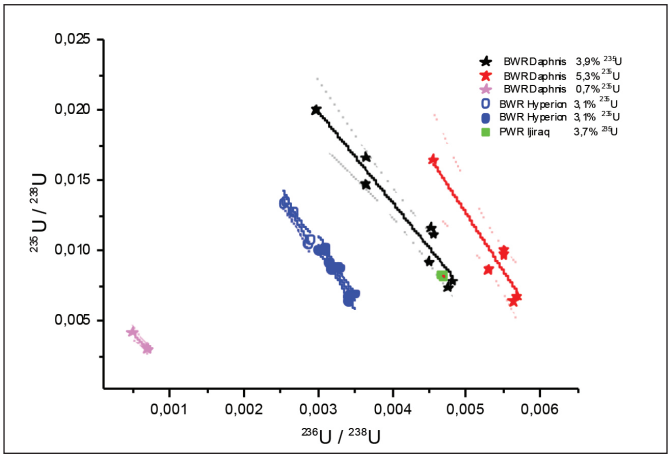
In order to better understand these graphs, Figure 1 was split in two separate graphs containing the samples of each reactor presented individually, as shown in Figure 3.

Figure 3 shows three different groups of samples in Daphnis, and two groups in the Hyperion data set, respectively.

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

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



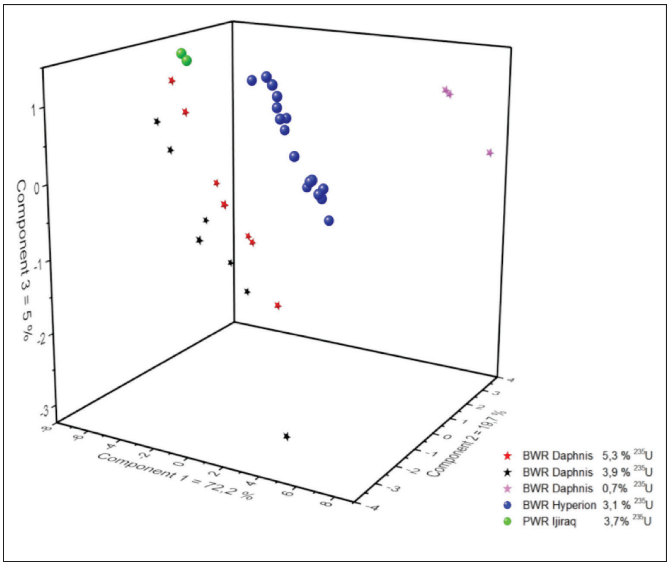
GWD/MT. Such a range of burnup values were likely due to exposure in different regions of the nuclear fuel assembly. Hyperion's samples have the same initial enrichment, but were separated in two groups with different burnup levels and plutonium profiles. There is a possibility that the samples may originate from the same fuel rod, but from different positions along the active length of the rod.

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.

Figure 5. PCA plots for the NNFL reactors



As shown, the PCA model was able to distinguish, with a high degree of confidence, each reactor as well as each class of samples.

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).

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

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

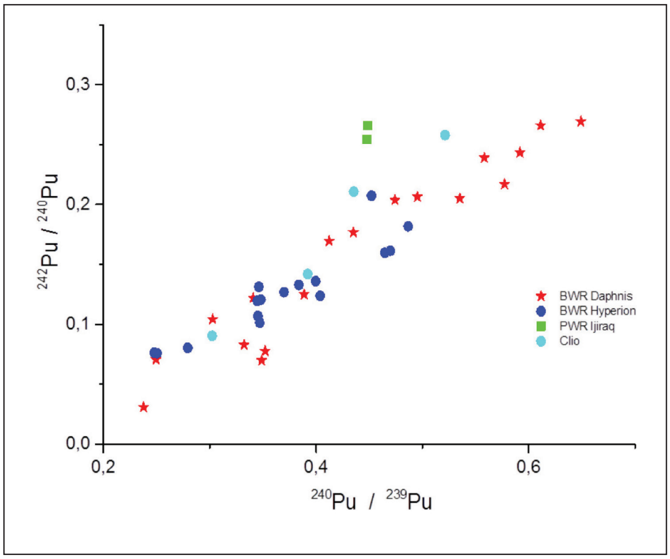


Figure 7. Isotope Correlation: $^{238}Pu/(^{239}Pu + ^{240}Pu)$ versus $^{240}Pu/^{239}Pu$ for NNFL reactors and Clio seizure

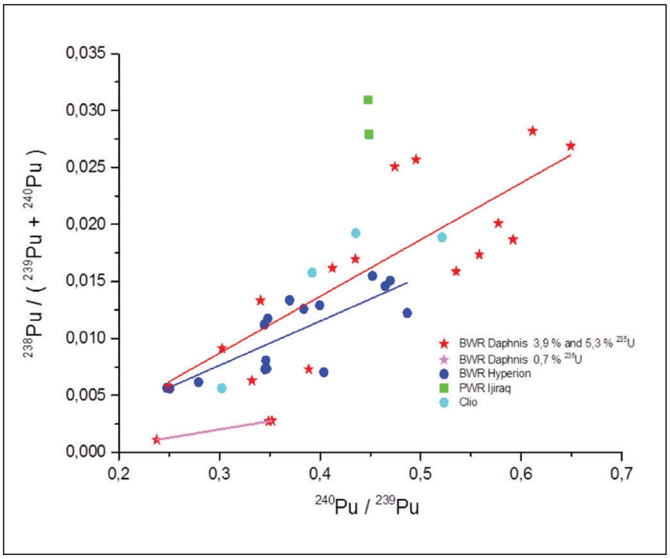


Figure 8. PCA of the NNFL's reactors and Clio

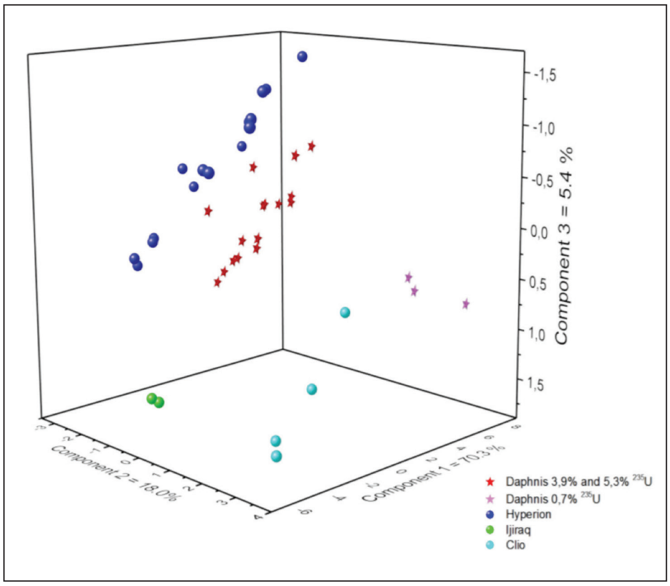


Table 1. Standardized coefficients for canonical variables

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

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.

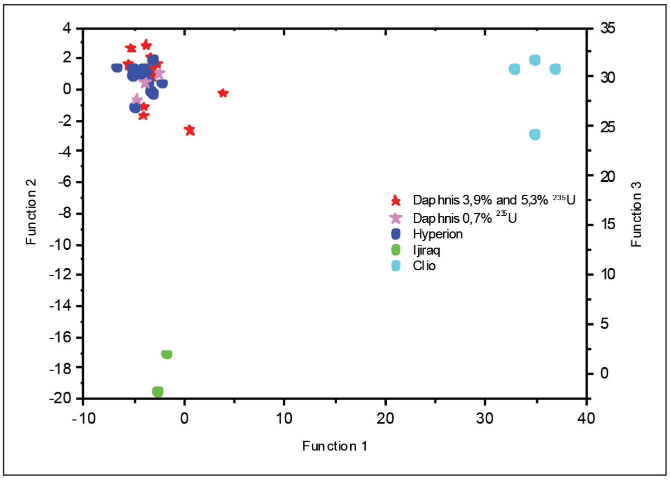
Conclusions and Lessons Learned

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 com-

Figure 9. Canonical distribution: NNFL and Clio sample



prised 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.

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Galaxy Serpent Virtual Tabletop Exercise: Canada's Approach, Findings, and Lessons Learned

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Abstract

Canada's participation in the Galaxy Serpent Virtual Tabletop Exercise (V-TTX) was aimed primarily at evaluating our current capabilities in the application of chemometrics (a subset of multivariate statistics) for the development of multistage pattern recognition algorithms for nuclear material attribution. To that end, Canada took a two-pronged approach in its library development and analysis methodology to determine whether or not a hypothetically seized spent nuclear fuel (SNF) sample originates from its assigned SNF inventory. The first approach involved the construction a spreadsheet-based library of the assigned SNF inventory isotopic measurements and ratios in a manner suited for the application of chemometrics. Here, measurements of the seized sample are interrogated and compared against inventory measurements using a multivariate statistics-based pattern recognition algorithm in order to establish an attribution probability. The second approach established a library of key isotopic correlations based on well-understood isotopic evolutions as a function of fuel burnup, against which similar correlations constructed from measurements of the seized sample are compared. The two approaches were applied independently of each other by two separate teams. Each team was able to conclusively attribute the seized sample to Canada's assigned SNF inventory. This paper examines the methods and results of each approach.

Introduction

Canada is currently undertaking a whole-of-government initiative to build a national nuclear forensics capability. A national nuclear forensics library (NNFL) was identified as a critical component of this capability. The Canadian Nuclear Safety Commission (CNSC) is charged with leading the development of Canada's NNFL. As part of its NNFL development activities, the CNSC led a team of subject matter experts (SMEs) from Atomic Energy of Canada Limited (AECL) and the National Research Council – Energy, Mines and Environment Portfolio (NRC-EME) for Canada's participation in the Galaxy Serpent V-TTX.

Canada has a well-established domestic capability in the application of chemometrics for the characterisation and attribution of nuclear material.[1,2] As such, chemometrics has been identified as the preferred approach in the development of a broader multistage pattern recognition algorithm for nuclear material attribution for use within Canada's NNFL. Canada's primary objective in participating in the Galaxy Serpent V-TTX was to evaluate its current chemometrics capability for nuclear material attribution.

In order to assess the chemometrics method and verify the results derived from it, Canada took a two-pronged approach in its participation in the Galaxy Serpent V-TTX. The library development and analysis methodology used to determine whether or not a hypothetically seized spent nuclear fuel (SNF) sample originates from its assigned SNF inventory included two distinct approaches undertaken independently by two separate teams. The first approach was undertaken by subject matter experts (SMEs) from the CNSC and NRC-EME, and consisted of constructing a spreadsheet-based library of the assigned SNF inventory isotopic measurements and ratios in a manner suited for the application of a multivariate statistics-based pattern recognition algorithm to interrogate and compare against measurements of the seized sample in order to establish an attribution probability. The second approach was undertaken by SMEs from AECL and consisted of establishing a library of key isotopic correlations based on well-understood isotopic evolutions as a function of nuclear fuel burnup, against which similar correlations derived from measurements of the seized sample were compared. The following sections discuss the technical details and results of each approach.

Spent Nuclear Fuel Inventory and Seized Sample Assigned to Canada

The SNF data assigned to Canada, and indeed all participants in the Galaxy Serpent V-TTX, was taken from the Spent Fuel Isotopic Composition Database (SFCOMPO), a public domain database of measured isotopic compositions of SNF, based on published literature, used for the validation of fuel burnup codes. SFCOMPO is operated and maintained by the Nuclear Energy Agency (NEA) Nuclear Science Division (NSD) of the Organization for Economic Cooperation and Development (OECD), under the supervision of the Working Party on Nuclear Criticality Safety (WPNCS).[3]

SFCOMPO catalogues the SNF isotopic composition from fourteen light water reactors (LWRs) from Germany, Italy, Japan, and the United States, including seven pressurized water reactors (PWRs) and seven boiling water reactors (BWRs). The SFCOMPO data characterizes SNF at four positions along the rod. The Galaxy Serpent V-TTX used SFCOMPO data that was augmented with measurement uncertainties. Each participant was assigned a measurement data set consisting of data from three nuclear reactors with assigned aliases, which represented a participant's SNF fuel inventory (i.e., holdings). Canada's assigned inventory consisted of SNF inventory data from three nuclear power plants (NPPs): Iapetus PWR, Janus BWR, and Mimas PWR. The assigned seized sample was given the alias Clio.

Chemometrics Approach to Attribution

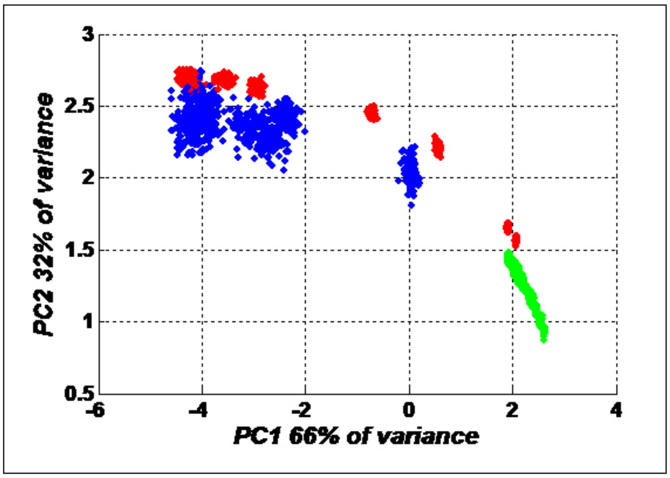
With the advent of sophisticated instrumentation, applied analytical chemistry has slowly evolved into a data-rich scientific field, thereby opening up the possibility of data-driven research.[4,5] Pattern recognition methods were originally designed to solve the class membership problem. In a typical pattern recognition study, samples are classified according to a specific spectral property or characterisation by using spectroscopic measurements. Other variables such as elemental composition, physical properties, color, and particle size (to name a few) can be used to resolve the class membership of an unknown sample of material. An empirical relationship (or classification rule) can be developed from a set of sample measurements for which the chemical compositions of interest are known (e.g., isotopic composition). The classification rule is then used to identify samples that are not part of the original training set. A training set consists of samples of a known class upon which a pattern recognition model can be built.

Principal component analysis (PCA) is a widely used multivariate analysis technique in science and engineering.[6] It is a method for transforming the original measurement variables into "new" uncorrelated latent variables (i.e., principal components). Each component is a linear combination of the original measurement variables. Using this procedure, a set of orthogonal axes that represent the direction of greatest variance in the data can be found. Variance for a data set is defined as the degree to which the data are spread in an *n*-dimensional measurement space. Typically, only two or three principal components are necessary to explain a significant fraction of the information present in multivariate data. Hence, PCA can be applied to multivariate data for the purpose of dimensionality reduction, the identification of outliers, the display of data structures and the classification of data points in an unsupervised manner. Under certain circumstances, the use of more advanced supervised classification methods, such as Soft Independent Modeling of Class Analogy (SIMCA),[7] which use a combination of multiple PCA and/or Partial Least Squares — Discriminant Analysis (PLS-DA)[6] methods, are necessary for the sorting of complex data sets. Supervised methods require a training set with known classifications in order to answer the specific question of attribution of an unknown sample. The use of the aforementioned techniques (PCA, PLS-DA, and SIMCA) were investigated for use to discriminate the populations of SNF from each NPP (i.e., class) within the SNF assigned inventory for the purpose of determining the provenance of the assigned seized sample Clio.

Building the Library and the Generation of Randomly Distributed Synthetic Data

In the application of pattern recognition algorithms, the number of variables that are significant (and necessary) to discriminate between different classes must be considered. First, it is important to have a sufficient number of data points to widen the variance for the dependent variables (i.e., isotopic ratios) in the data set. Typically, the number of calibration data points should be larger than the number of dependent variables.

Figure 1. PCA of the library 4600 fuel rods ("synthetic data") for ninety-one variables with 50 percent missing values (Red: Mimas; Blue: Iapetus; Green: Janus)



Since the multivariate approach needs more samples than variables, a "synthetic" SNF inventory database was generated using the uncertainties provided for each value in the assigned fuel inventory. Since uncertainties are provided for 1σ standard deviation for each measurement (data point), it is possible to generate randomly distributed measurements around the average measurement value. As such, for each fuel rod data point, synthetic values were generated by calculating the average value ± 2.5σ standard deviations (i.e., 99 percent of the normalized Gaussian distribution). Hence, 100 synthetic measurement data points were generated for each fuel rod in the assigned SNF inventory, whereby the forty-six fuel rods in the assigned inventory were transformed into a database (or library) of 4,600 fuel rods with ninety-one measurement variables.

Chemometrics Analysis Results

PCA was used on the synthetic library as an exploratory analysis, the results of which are presented in Figure 1. The formation of a cloud around each individual fuel rod can be observed. More importantly, however, the application of PCA with two components can almost entirely resolve the SNF inventory in the library with a high degree of discrimination. From this, it is possible to conclude that more advance supervised models such as SIMCA or PLS-DA could provide a more refined resolution to the pattern recognition problem.

Although PCA has demonstrated good selectivity for classifying the different NPPs, its unsupervised nature makes it less convenient for training and the discrimination of the unknown seized sample Clio. Consequently, it was decided to use supervised techniques to more thoroughly interrogate the measurement data in the library.

A preliminary assessment of SIMCA-derived results (not shown) reveals that SIMCA does not adequately discriminate the different classes of the assigned SNF inventory. PLS-DA, however, was successfully used to resolve the class membership between the different fuel rods from the three NPPs. Because several individual measurements for several rods did not have a value, it was decided to remove variables for which there were multiple missing measurement values. Thus, the PLS-DA model was built using only seven out of the twelve discrimant variables (i.e., 40 percent missing values, Figure 2). These seven variables include: ^{137}Cs/^{238}U, ^{235}U/^{238}U, ^{236}U/^{238}U, ^{238}Pu/^{238}U, ^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu and ^{242}Pu/^{239}Pu.

The results of the PSL-DA model shown in Figure 2 have been pre-treated by removing the average variable value and normalizing the variance to a value of one. The model was then cross-validated using randomized data subsets with a block size of 460 data points for five iterations. The model prediction for the Iapetus class inventory is shown in Figure 2a. It is observed that the Iapetus measurement data points lie above the attribution probability threshold (0.45), which is determined by Bayesian statistics, whereas the measurement data points for Janus and Mimas lie below the threshold. This indicates that measurement data points for Iapetus can be wholly discriminated from those of Janus and Mimas. In addition, it is observed that the measurement data points of the seized samples for Clio (Clio-1, Clio-2, Clio-3, and Clio-4) also lie above the model attribution probability threshold for Iapetus, which indicates a conclusive positive determination that all four seized Clio samples can be attributed to the Iapetus class. In order to determine whether or not the Clio attribution is unique to Iapetus, the PLS-DA analysis must be repeated using the Janus and Mimas measurement data points.

The prediction obtained by the PSL-DA model for Janus is presented in Figure 2b, with a model attribution probability threshold of 0.65. Here, all the measurement data points of fuel discharged from Janus have a class membership probability higher than 0.65, which confirms that they belong to Janus. In addition, the measurement data points from Iapetus and Mimas, as well as those of the seized sample Clio, lie below the class membership threshold, indicating that they are not members of the Janus class. The model predictions for the Mimas are presented in Figure 2c, show identical results to Janus in that

Figure 2. Analysis of the 4600 fuel rod synthetic database for 40 percent of missing variable values by PLS-DA. The 400 fuel rod measurements of the seized material Clio have been added to test and validate the PLS-DA model.

Figure 2a. PLS-DA model prediction for Iapetus

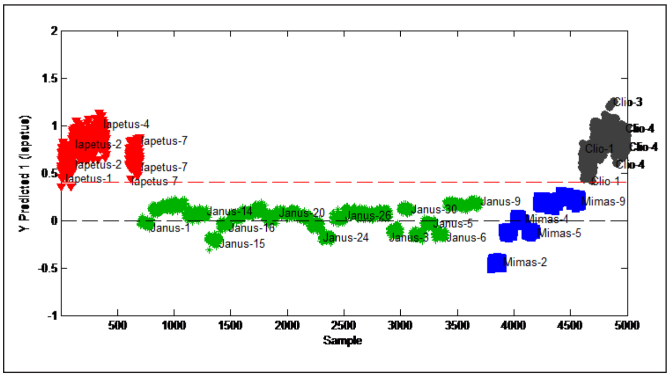


Figure 2b. PLS-DA model prediction for Janus

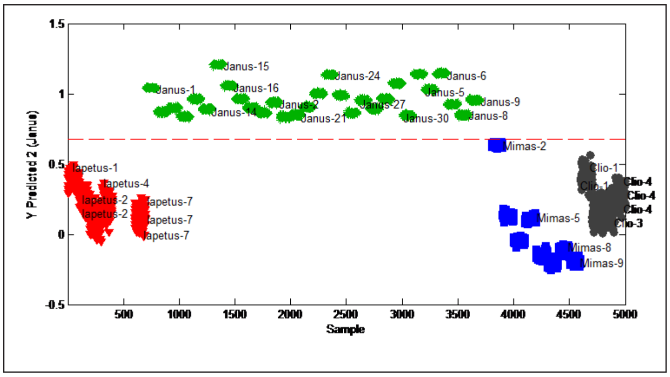
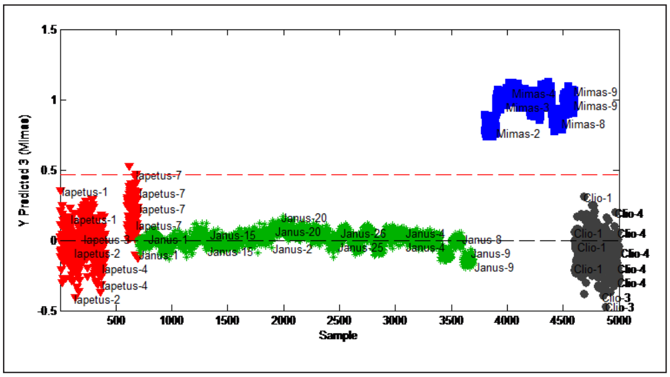


Figure 2c. PLS-DA model prediction for Mimas



Mimas measurement data points lie above the membership threshold of 0.48, with Janus and Iapetus class measurement data points lying below. In addition, the Clio measurement data points also lie below the class membership threshold, thus excluding them from being of Mimas provenance.

Conclusions Derived from the Chemometrics Approach to Attribution

It is concluded that all four seized Clio samples originate from fuel discharged from the Iapetus PWR NPP.

The generation of synthetic normally distributed measurement data using a 1σ uncertainty (that was provided with each measurement value) does not affect the fundamental ability of chemometrics models to cluster and decompose the measurement data using the inherent relational characteristics of the data. In addition, an analysis of the model loadings has revealed that discrimination of the measurement data is governed primarily by plutonium and uranium isotopes variables, which was independently confirmed in the isotopic correlations approach discussed in the following sections.

The results of the chemometrics analysis are very encouraging in that they were confirmed using the more traditional isotopic correlations approach in analysing this type of subject matter data. More importantly, the chemometrics approach can be taken independent of subject matter knowledge as it is not required to build the necessary classification rules to discriminate the classes of the measurement data set.

Isotopic Correlations Approach to Attribution

Building the Library

The first step in the isotopic correlations approach was to compile the assigned SNF inventory data. The inventory data were examined to identify any inconsistencies or errors. It was observed that sample numbers were absent in the original data set. This observation was communicated to the V-TTX organizers; and as a result, the data sets were revised to include sample numbers.

The second step was to explore the applicable isotopic correlations, starting with the eighteen correlations provided in the technical guide for the V-TTX, in addition to using other isotopic ratios reported in the literature.[8,9] The objective in exploring the different isotopic correlations was to determine if any of them can provide distinct trends (i.e., that can be used as signatures) which would be useful in the attribution of Clio. Several isotopic correlations appeared to be important in distinguishing among measurements taken from the three reactors. See Table 1.

Table 1. Isotopic correlations from the three reactors

Burnup vs. ^{235}U	^{238}Pu/^{238}U vs. Burnup
^{234}U vs. ^{235}U	^{241}Pu/^{239}Pu vs. Burnup

^{236}U vs. ^{235}U	^{240}Pu/^{239}Pu vs. ^{235}U
^{234}U vs. ^{236}U	^{242}Pu/^{240}Pu vs. ^{235}U
Pu_{total}/^{238}U vs. ^{235}U	^{242}Pu/^{238}Pu vs. ^{242}Pu/^{239}Pu
^{239}Pu/^{238}U vs. Burnup	

Isotopic Correlation Analysis for the Attribution of Clio

The measurements for Clio were compared with the isotopic correlations listed above. The results of the isotopic correlation analysis are discussed first as correlations based on uranium isotopes and, then as correlations based on plutonium isotopes.

Correlations Based on Uranium Isotopes

As shown in the plot of burnup vs. ^{235}U (Figure 3), the amount of ^{235}U declines logarithmically with burnup because of increasing ^{239}Pu fission. This correlation may be used to estimate the starting enrichment of the fresh fuel (i.e., at zero burnup).

Figure 4 shows that the amount of both ^{234}U and ^{235}U decline logarithmically with burnup. The ratio expected to fit ^{234}U/U_{total} to ^{235}U/U_{total} is strictly logarithmic, but is very close to being linear. Extrapolating ^{234}U vs. ^{235}U back to the initial ^{235}U content indicates that the Janus fuel and Iapetus fuel had different initial ^{234}U content. The higher ^{234}U content in Iapetus is consistent with fuel that has been reprocessed.

Lastly, ^{236}U builds up linearly with ^{235}U depletion. The correlation shown in Figure 5 indicates a non-zero initial value for ^{236}U, which is consistent with fuel that has been reprocessed.

The above-mentioned correlations based on uranium isotopes indicate that the Clio seized sample is consistent with fuel discharged from the Iapetus PWR NPP.

Figure 3. Burnup vs. ^{235}U/U_{total}

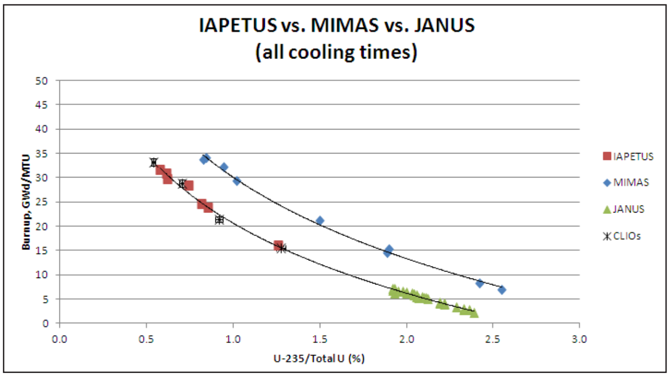
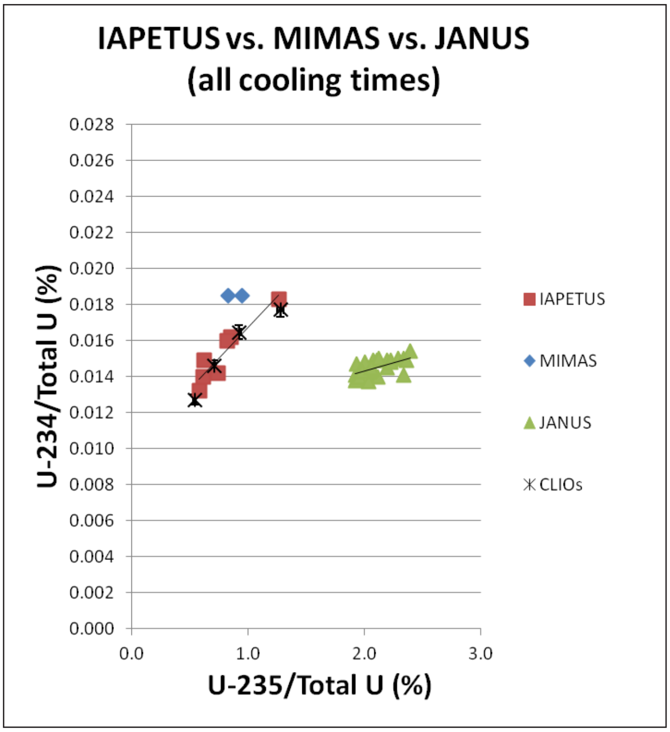


Figure 4. ^{234}U/U_{total} vs. ^{235}U/U_{total}



Correlations Based on Plutonium Isotopes

Figure 6 shows the correlations between Pu_{total}/^{238}U vs. ^{235}U. As can be seen, there is one Iapetus measurement data point (Iapetus-7) that appears to be inconsistent with the remainder of the Iapetus measurement data cluster. It is believed that this inconsistency is due to a lower-than-normal value of ^{238}U. By correcting this data point (using the theoretical burnup value), the scatter among the Iapetus measurement data points is reduced.

As expected, the correlation between ^{238}Pu/^{238}U vs. burnup (Figure 7) shows that ^{239}Pu build-up is saturating at higher burnup values. This correlation shows that Clio seized sample measurements are most similar to Iapetus measurements.

In the correlation between ^{238}Pu/^{238}U vs. burnup (Figure 8), there is another Iapetus measurement data point (Iapetus-2) that appears to be inconsistent with the remainder of the Iapetus measurement data cluster. It is believed that this inconsistency is due to a higher-than-normal value of ^{238}Pu. There is no separation of the reactors in this correlation. If all reactors are considered to lie on the same fit, then Iapetus-2 can be seen to have ~50 percent too much ^{238}Pu. By correcting this data point, the scatter among the Iapetus measurement data points is reduced. It is noted that one of the Clio measurement data points seems to mimic the Iapetus-2 anomaly observed for ^{238}Pu. If the anomaly is a real effect, then this would be a powerful indicator that Clio is consistent with Iapetus.

Figure 5. $^{236}\text{U}/\text{U}_{total}$ vs. $^{235}\text{U}/\text{U}_{total}$

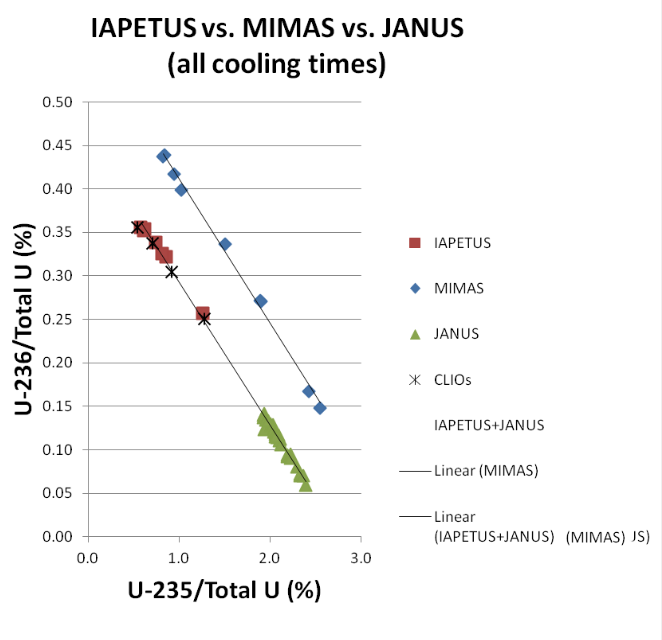


Figure 6. $\text{Pu}_{total}/^{238}\text{U}$ vs. $^{235}\text{U}/\text{U}_{total}$

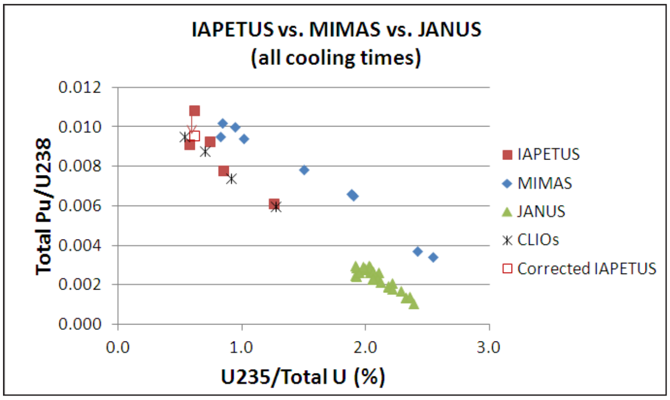


Figure 7. $^{239}\text{Pu}/^{238}\text{U}$ vs. Burnup

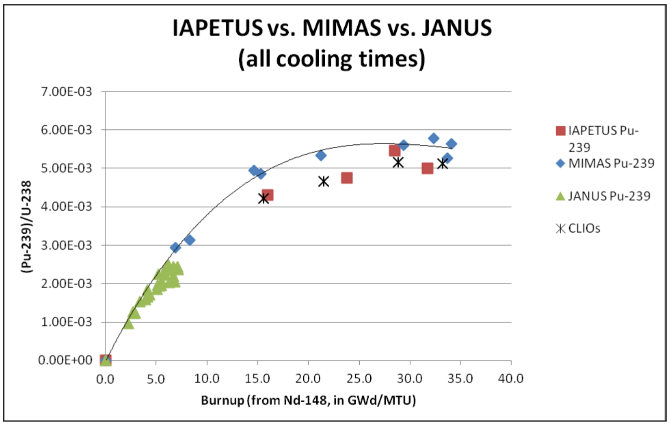


Figure 8. $^{238}\text{Pu}/^{238}\text{U}$ vs. Burnup

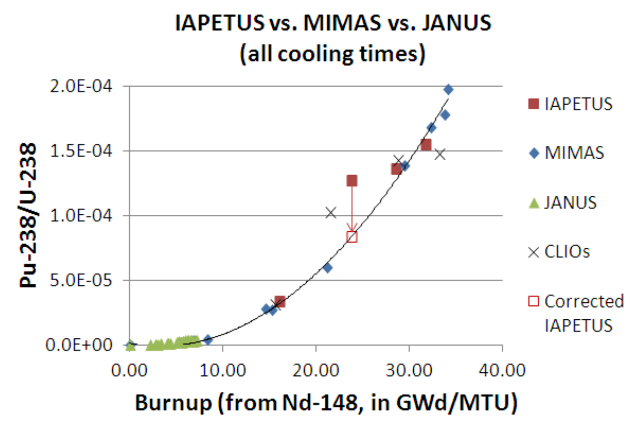
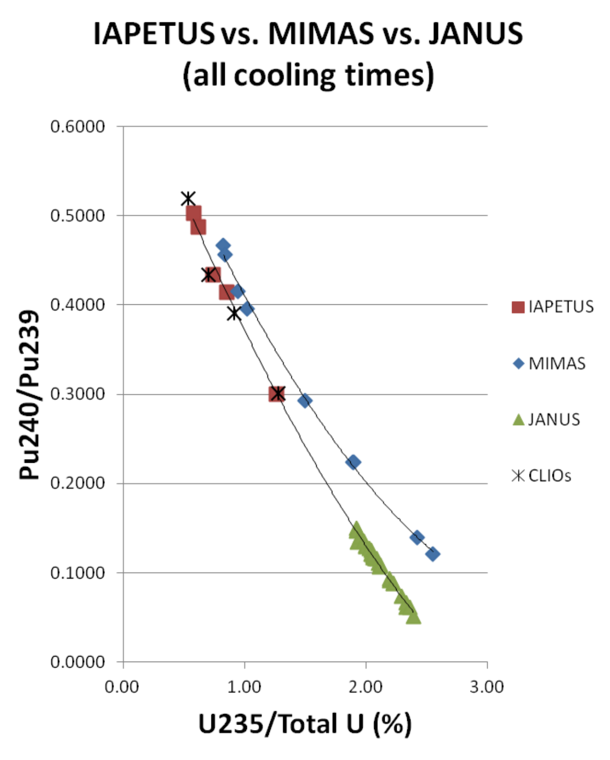


Figure 9. $^{240}\text{Pu}/^{239}\text{Pu}$ vs. $^{235}\text{U}/\text{U}_{total}$



The ratio of $^{240}\text{Pu}/^{239}\text{Pu}$ is expected to increase linearly with decreasing ^{235}U once ^{239}Pu reaches equilibrium (Figure 9). In addition, it is recognized that ^{239}Pu reaches equilibrium faster for lower initial ^{235}U enrichment.

The correlation between $^{242}\text{Pu}/^{238}\text{Pu}$ vs. $^{240}\text{Pu}/^{239}\text{Pu}$ (Figure 10) is reported in the literature[8,9] as being able to differentiate reactors by both neutron spectrum and specific power. The

Figure 10. ^{242}Pu/^{238}Pu vs. ^{240}Pu/^{239}Pu

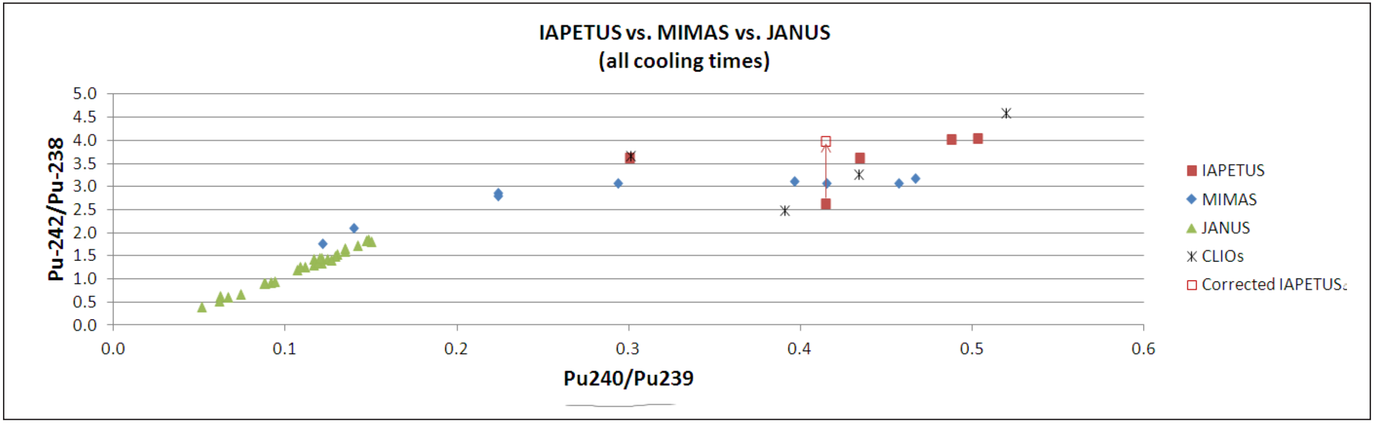
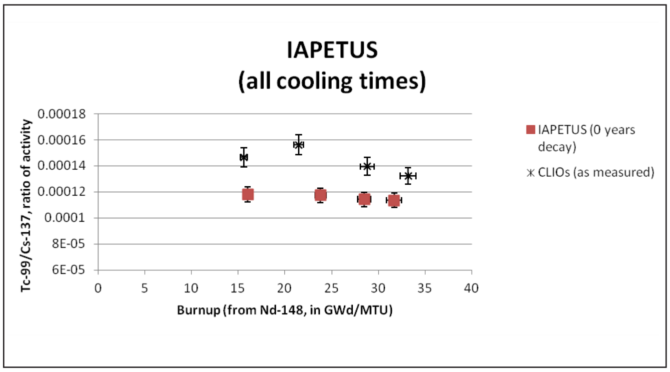


Figure 11. ^{99}Tc/^{137}Cs (ratio of activity) vs. Burnup



^{238}Pu anomaly seen here is observed in both the Iapetus and Clio measurements.

The above-mentioned correlations based on plutonium isotopes indicate that Clio seized sample is consistent with fuel discharged from the Iapetus PWR NPP.

Conclusions Derived from Correlation Based Isotopic Analysis

An analysis using the isotopic correlations discussed earlier is sufficient to make a conclusive positive determination that Clio is attributable to fuel discharged from Iapetus.

Clio Age Dating Based on Fission Products and Plutonium Isotopes

The age of Clio (i.e., time after discharged from the reactor) can be determined using the following technique:

1. Perform the decay correction of the Iapetus measurements to $t = 0$, i.e., based on the known age of the Iapetus samples, calculate the abundances of the various fission products or actinides at the time the Iapetus samples were discharged from the reactor.
2. Fit a curve to the decay-corrected ($t = 0$) Iapetus measurements vs. burnup.
3. Estimate the age of Clio by:
    a. Performing the decay correction of the Clio measurements (similar technique to decay-correcting the Iapetus measurements) to an age where the square of the deviations of the decay-corrected Clio measurements from the decay-corrected Iapetus curve is minimized; or
    b. Determining the age of each individual Clio measurement (i.e., calculating the decay time that makes each Clio measurement coincide with the decay-corrected Iapetus curve), and taking the average age.

As shown in Figure 11 and Figure 12, Iapetus and Clio measurements can be fitted onto a plot consistent with the ratio of ^{99}Tc/^{137}Cs vs. burnup by "un-aging" the Clio measurements. Using method 3a described above, Clio's age was estimated at 9.4 ± 3 years; and using method 3b, its age was estimated at 9.2 ± 3 years.

An alternative measurement that can be used for age dating Clio is the ratio of ^{240}Pu/^{241}Pu. As shown in Figure 13 and Figure 14, by plotting the ^{240}Pu/^{241}Pu ratio vs. ^{235}U enrichment, and by "un-aging" the Clio measurements, Clio appears to be consistent with fuel discharged from Iapetus. Using the ^{240}Pu/^{241}Pu ratio, both methods 3a and 3b as described yield a Clio age estimate of 10.4 ± 0.4 years, consistent with the result obtained using ^{99}Tc/^{137}Cs measurement.

Based on the age dating methods described above, the relative abundances of the examined radioisotopes in Clio lead

Figure 12. ^{99}Tc/^{137}Cs (ratio of activity) vs. Burnup with the decay of Clio samples taken into account

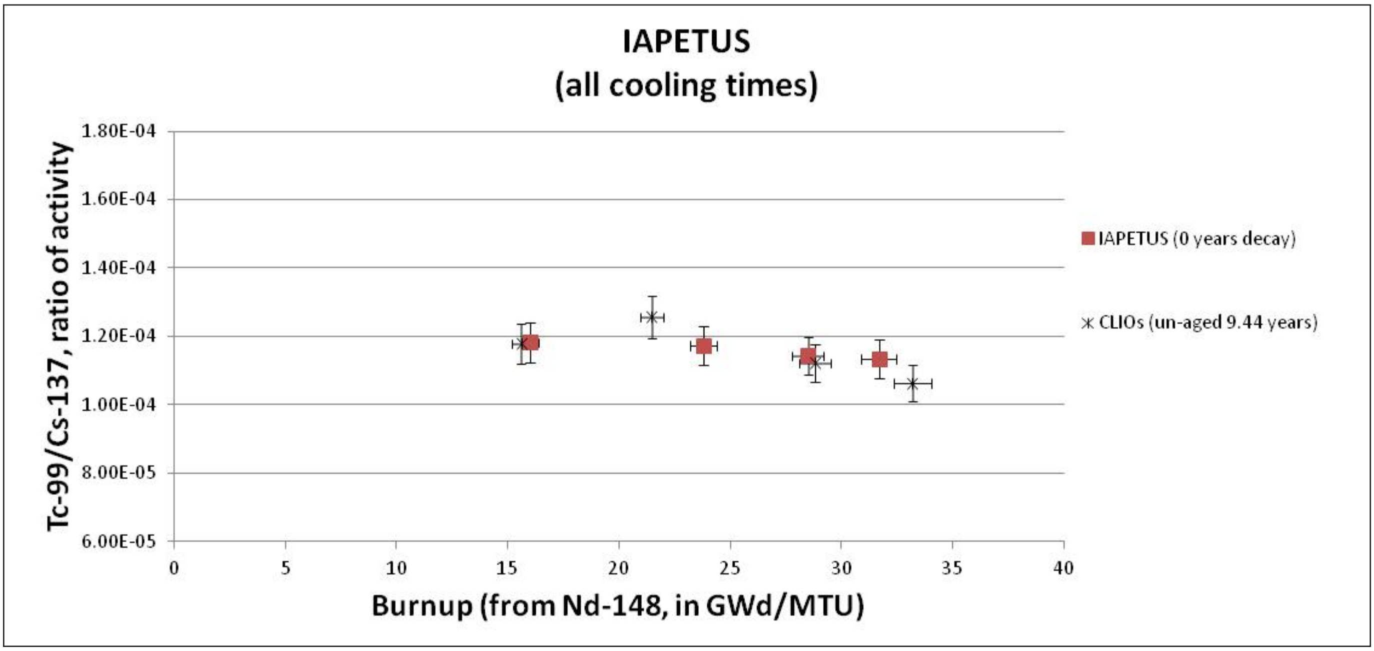


Figure 13. ^{241}Pu/^{240}Pu vs. ^{235}U/U_{total}

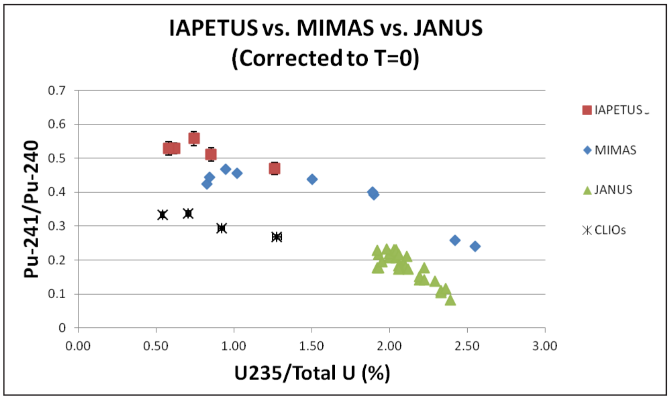
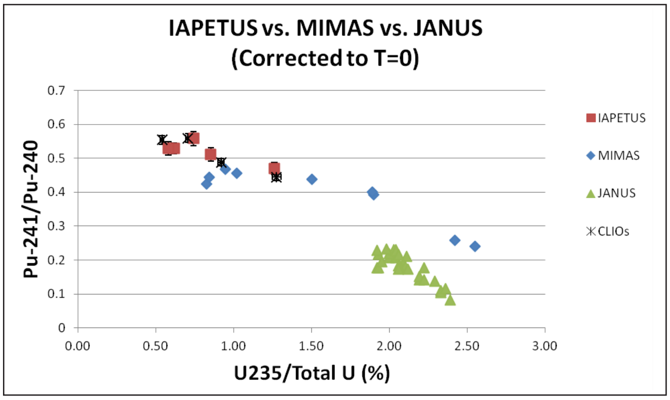


Figure 14. ^{241}Pu/^{240}Pu vs. ^{235}U/U_{total} (decay of Clio samples taken into account)



to a determination that the fuel rod from which Clio originates has been out of the reactor for approximately ten years.

Conclusion

Canada identified the Clio seized samples originate from fuel discharged from the SNF inventory belonging to the Iapetus NPP. This was achieved using two approaches carried out independently by two separate teams.

Lessons Learned

Galaxy Serpent V-TTX highlighted key lessons learned regarding the practice of material attribution through analytical means. These are:

- The use of the chemometrics (multivariate statistics) approach is a powerful method to decompose and discriminate data using inherent relational characteristics. This approach can be taken with relatively little subject matter expertise of the material in question and was shown to tolerate significant variance and deficiencies in the measurement data.
- For the specific application of the Galaxy Serpent V-TTX subject matter, the support of nuclear fuel and reactor physics SMEs to assist in quickly identifying the key relationships within the measurement data set accelerated the development of chemometrics-based models.
- The use of isotopic correlations by relevant SMEs allowed for the identification of inconsistencies in the measurement

data. This underscores the importance of validating the quality of the data that is to be incorporated into a NNFL.
- Spent fuel data analysis and interrogation is supported by an extensive and robust body of work describing the appropriate methods. This perhaps circumvented the need to build a library given that the key relationships that needed to be extracted from the data are well-understood.

Ali El-Jaby, PhD, P.Eng., obtained his PhD in nuclear engineering from the Royal Military College of Canada, where his studies focused on the development of nuclear fuel behavior models. Immediately after his PhD studies, El-Jaby joined the CNSC, working in the areas of reactor and nuclear fuel safety, emergency response operations, nuclear material safeguards and forensics. Currently, El-Jaby is leading the CNSC's nuclear forensics activities and the development of Canada's NNFL.

Having obtained his Nuclear M.Eng. from the Royal Military College of Canada, Rick Kosierb has spent more than twenty years in the areas of radiation detection instrumentation, protective clothing and decontamination. His Canadian government dealings in nuclear forensics and to find innovative detection equipment for the International Atomic Energy Agency led him to the Chemometrics and Laser-Induced Breakdown Spectroscopy scientific fields.

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Geoffrey Edwards came to reactor physics at AECL in 1993 from a background in high-energy scattering physics. His career has included time as the NRU Duty Physicist and the commissioning of the Slowpoke and MAPLE reactors. He is currently chair of the Nuclear Criticality Safety Panel at AECL.

Duncan Barber, PhD, is a chemist and nuclear engineer with Fuel and Fuel Channel Safety at AECL. His current research interests include fission product release from fuel, radionuclide generation and depletion, and thermodynamics of nuclear fuel.

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Lessons Learned from the International Tabletop Exercise of National Nuclear Forensics Library at JAEA

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Abstract

The Japan Atomic Energy Agency (JAEA) initiated an R&D project focused on the maturation of nuclear forensics technology from 2011. This project includes the development of a prototype national nuclear forensics library (NNFL), and the JAEA participated in the first international tabletop exercise of NNFL Galaxy Serpent, held by the International Technical Working Group as a part of our NNFL development project. In this paper, the JAEA investigation during the tabletop exercise is described and the information gathered from the experience is discussed in detail: A model NNFL of spent nuclear fuels (SNFs) including one boiling water reactor (BWR) and two pressurized water reactors (PWR-1 and PWR-2) was completed in the first phase of the exercise. In the second phase, by using isotope correlation comparison technique and principal component analysis it was found that a seized nuclear material was definitely consistent with the SNF of PWR-2. Furthermore, the lessons learned from these exercises will be effectively applied to the further development of the NNFL at JAEA. Finally, we include a brief update on the current status of the NNFL development project at JAEA.

Introduction

Since the early 1990s, there has been increasing international concern regarding the illicit trafficking of nuclear or other radioactive materials. By 2011, over 2,000 illicit trafficking cases had been reported to International Atomic Energy Agency (IAEA).[1] Once the unknown nuclear/radioactive materials are detected and seized from a nuclear security event, questions regarding their origin, history, and intended use must be addressed. These questions can be answered using nuclear forensic tools, which also help to identify deficiencies in the national nuclear security system.[2] Recently, the consolidation of nuclear forensic capabilities such as analysis technologies and national frameworks has become a significant issue for many countries that wish to strengthen their national nuclear security measures through the development of their national capabilities.

In November 2009, the Japanese and United States governments signed the "Japan-U.S. Joint Statement Toward a World Without Nuclear Weapons" at the U.S.-Japan summit meeting.[3] In this statement, it was declared that both governments agreed to nuclear nonproliferation, the adoption of safeguards, and security cooperation. Such an endeavor involves the use of nuclear forensics in addition to measurement and detection technologies, human resource development, training, and infrastructure assistance for countries interested in nuclear energy, and the coordination of respective member states support programs for IAEA safeguards. Furthermore, at the Nuclear Security Summit in 2010 (Washington, DC, USA), the Japanese government issued a national statement encouraging the development of forensic technologies related to the measurement and detection of nuclear materials within a three-year timeframe. The technologies developed as a result of this effort were to be shared with the international community as a means of strengthening the global nuclear security system.[4] In response to these two statements given by the Japanese government, the Japan Atomic Energy Agency (JAEA) has initiated an R&D project in 2011 to develop nuclear forensics technologies.

The R&D project at JAEA covers nuclear forensics analysis technologies such as isotope analysis, trace element measurement, age determination, and particle analysis; the development of a national nuclear forensics library (NNFL) prototype is also included. The goal of this first step in the JAEA's NNFL development project is to study the concept of an NNFL for Japan and, subsequently, to develop a nuclear materials database prototype based on the conceptual study. We expect to reach this goal by the end of the first quarter of 2014. As a part of the development project, the JAEA participated in Galaxy Serpent, an international tabletop exercise on NNFL. In this paper, JAEA's experiences with the Galaxy Serpent exercise are described and the information gathered from this interaction is discussed. The tabletop exercise proved to be a useful tool for learning the overall process of creating an NNFL, as it involves library construction based on existing raw data of nuclear materials and continues to the analysis of seized nuclear materials and then the final reporting of the conclusion reached by comparison of the analyzed data with the library database. An update on the current status of the NNFL at JAEA is also provided in this paper and it includes an account of the lessons learned from the agency's experience with the *Galaxy Serpent* exercise.

Galaxy Serpent Tabletop Exercise

Galaxy Serpent is the first entirely virtual, web-based international tabletop exercise advancing the concept of NNFL. The exercise was conducted under the auspices of the Nuclear Forensics International Technical Working Group (ITWG). Details of the Galaxy Serpent are given in a separate paper.[5] The JAEA participated in the second round of the exercise as the only participating organization from Japan. The JAEA team included eight staff members with backgrounds in analytical chemistry, reactor physics, safeguards, nuclear security, and computer engineering. In the present exercise, spent nuclear fuel (SNF) isotopic compositions obtained by post-irradiation examination were used as the nuclear material data for the hypothetical NNFL and seizure.[5] Thus, expertise in the fields of analytical chemistry and nuclear engineering were key to analyzing the seizures in this exercise.

Each team in this exercise was given a unique galaxy name to provide anonymity, and given SNF data from hypothetical nuclear reactors assumed to represent all nuclear and radioactive material holdings in the given galaxy.[5] The names of our team and the analyzed reactors and seizures are given in Table 1. The name of the representative galaxy given to the JAEA team was *URSA* and the isotopic compositions of SNFs derived from three different nuclear reactors were provided as raw data for the creation of our own NNFL. Our group of reactors was composed of one boiling water reactor (BWR) and two pressurized water reactors (PWRs), and the SNF data included the isotopic compositions of uranium, plutonium, and other transuranic elements and fission products, the compositions of which depended on each burnup step. Various types (units) of data were provided for the isotope compositions along with their associated uncertainties. The differences between the data for different reactors could not be easily identified because the isotope compositions were measured using different burnup steps for each reactor. In addition, the data units used for each measurement also differed among the three reactors. One way to overcome this issue is to unify their different data into a same unit, plot the unified data as a function of isotope ratio, relative weight or burnup, and then compare their isotope correlations for evaluation. After our NNFL database was completed by converting the SNF data into those with same unit and plotting their isotope correlations, the isotope composition data for a hypothetical seizure was provided by the exercise organizers in a similar format to that of the SNF data used earlier in the exercise.

The second round of the tabletop exercise was initiated at the beginning of April 2013 and took approximately three months for the JAEA to finish (Table 2). The exercise was completed in two phases: NNFL creation (phase 1) and seizure analysis (phase 2). In particular, the aim of phase 2 was to determine whether a seizure was or was not consistent with materials registered in the model NNFL. The actual working time for each phase was approximately two to three weeks with 20 man-h per week allocated to the exercise. The exercise took longer to complete than the time estimated by the organizers[5] because a significant amount of time was spent determining differences in composition trends for each of the three reactors as well as identifying the respective burnup characteristics following the NNFL creation. Ultimately, we were able to confirm with the organizers that our conclusions regarding the seizure and the registered materials in the NNFL were correct. Methodologies, detailed results of the exercise, and discussions are described in the following sections.

Results and Experiences

An NNFL is primarily used as a tool to assess if a nuclear/radioactive material encountered out of regulatory control is consistent with the materials produced, used, or stored within a country.[6] It may consist of a material database, an associated system for data handling, and other analyzing systems such as multivariate statistical analysis and image analysis. All nuclear/radioactive materials should ideally be registered in the NNFL. The size and format of the material database highly depends on the scale of the nuclear fuel cycle and the amount and complexity of materials existing from the past to the present in a country. Because the amount of data we needed to handle was small and relatively simple, our NNFL for the *URSA* galaxy was created in the form of a spreadsheet using Microsoft Excel. If the amount of given data is much larger or more complex in nature, other computational tools such as a relational database could be used to form a material database. The differences between SNF isotope compositions in different nuclear reactors were studied using isotope correlation comparison after the NNFL was created. The URSA galaxy was assumed to encompass one BWR and two PWRs. In general, there are some differences between the core characteristics of BWR and PWR that affect the isotope compositions produced in the SNFs. In this exercise, the differences in SNF isotope compositions between the BWR and PWRs were easily determined with the given SNF data. On the contrary, it was very difficult to precisely determine the differences between the two PWRs (Mimas and Siarnaq). The characteristic plots of the isotope correlations that showed some differences between the two PWRs were summarized and compiled as a graphical plot database to be utilized for seizure analysis in the next phase of the exercise.

In phase 2, two different approaches were used to assess if the seizure was consistent with materials registered in the *URSA*-NNFL: isotope correlation comparisons and principal component analysis (PCA). Isotope correlations of the seizure Erato were examined based on the degree of coincidence, which was defined for convenience of classification (Table 3). The degree of coincidence was evaluated by visual estimation as shown in Figure 1. An example of our seizure analysis by isotope correlation comparison is listed in Table 4. It was shown that the seizure was not consistent with the SNF derived from the BWR. In contrast, the other two PWRs showed similar isotope correlation trends with the seized material. As described previously, it was extremely difficult to distinguish between the two PWRs and, consequently, additional comparisons were necessary for forty-five combinations of isotope correlations. Table 5 shows the summary of these additional comparisons. From these comparisons, we see that *PWR-2* (Siarnaq) is more consistent than PWR-1 (Mimas) for 25/45 items and, furthermore, it is more consistent for 11/20 items that were categorized in the same comparative grade (namely, the pairs of A-A, B-B, and C-C for the degree of coincidence in Table 3). As a result, about 80 percent of the compared combinations showed maximum consistency for PWR-2, and therefore, it was strongly concluded that the seized material is associated with the PWR-2 reactor with an 80 percent confidence level. PWR-1 also shows some consistency; however, there was no corroboration using isotope correlation, which definitely indicates a stronger consistency with PWR-2.

Figure 1. Example of isotope correlation plot (^{235}U/Total-U vs. ^{242}Pu/^{240}Pu)

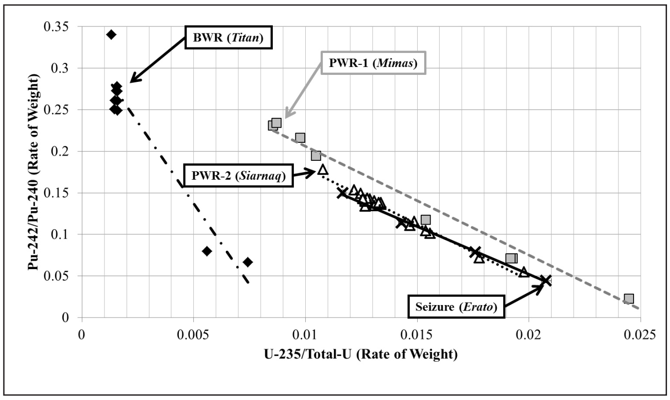
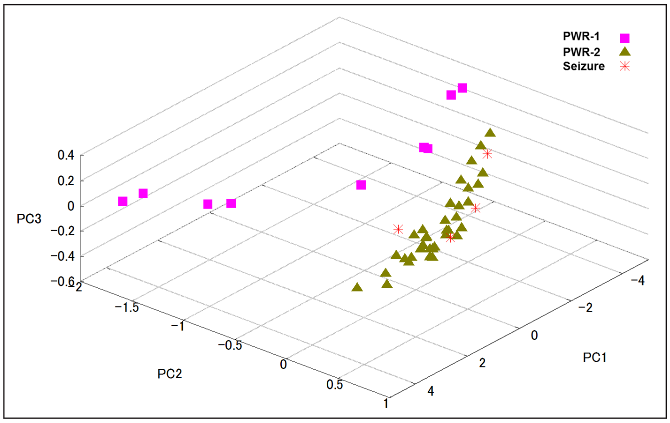


Figure 2. PCA score of plutonium isotope compositions (Explanatory variable of ^{238}Pu (%), ^{239}Pu (%), ^{240}Pu (%), ^{241}Pu (%), ^{242}Pu (%) were used)



As an alternative approach, we also applied the PCA method, a widely used multivariate statistical procedure, to our seizure analysis. While the isotope correlation method is limited in the number of comparable isotope compositions in this exercise, the PCA method can allow for the assessment of many items simultaneously. The isotope compositions of the seized material and the two PWRs were examined using the PCA. An example of a PCA made for plutonium isotope compositions is shown in Figure 2. The differences between the two PWRs are obvious from the PCA results, and it was confirmed that the seized material is more consistent with PWR-2 than PWR-1. Therefore, the results derived from the isotopic correlation comparison method can be verified by the PCA method and together led to a conclusive positive finding that the seizure originated from PWR-2, a finding which was verified by the exercise organizers.

Technical Lessons Learned and Their Application to NNFL Development

Some technical lessons for NNFL development at the JAEA learned from its experience with this tabletop exercise are the following:

- processing of raw material data into a library database,
- identification of the key characteristics of registered materials,
- expertise for seizure analysis or knowhow on data utilization in an NNFL,
- quantitative measure in comparative analysis, and
- reliability or confidence level regarding the results of the seizure analysis.

Compiling a material database is one of the most important issues in NNFL development, and the majority of compilation is the processing of raw material data into a standard format fitted for the database. In many cases, the raw material data to be registered in the NNFL comes in a variety of formats (e.g., electronic/hard copy, various units). The data provided in the present tabletop exercise varied significantly for each reactor even though only the SNF isotope composition data was provided. In most cases, the amount of raw material data collected for an NNFL should be larger and more complicated in nature, thereby making the task of data processing far more difficult and time consuming. To ensure successful data processing and database compiling, it is necessary to perform a preliminary survey on existing materials. The design of a nuclear material database in an NNFL depends strongly on the nuclear fuel cycle, which refers to the amount and variety of the materials available. A good preliminary survey makes it possible to know the variety and amount of target materials, relevant data, and other related information. It is also helpful to establish a clear vision of the scale and complexity of the material database. The standard data format for the material database should be defined by its specific design, which is based on the preliminary survey. It is also helpful to develop guidelines regarding the task of data processing and compilation into the database (the procedures and the data priority, for example).

Figure 3. Design outline of the NNFL developed at JAEA[7]

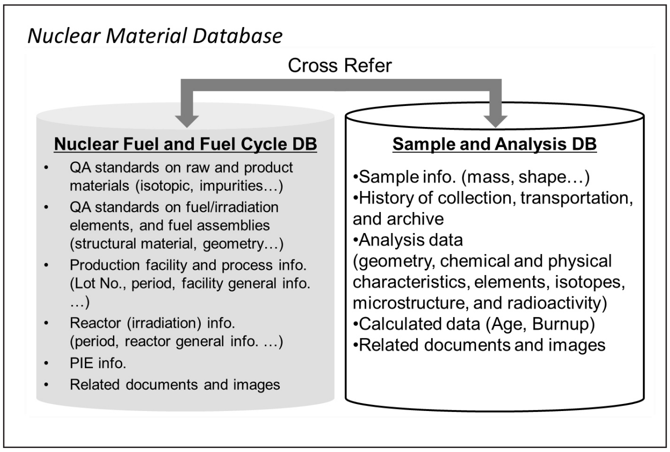
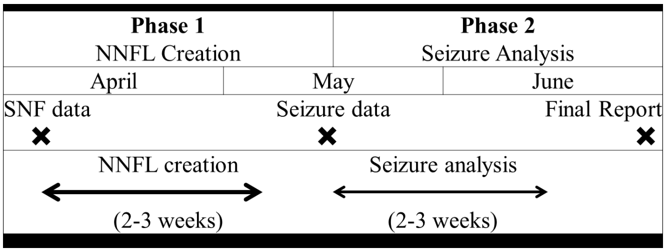


Table 1. Names allocated in the exercise

Round	second
Galaxy name (team name)	*URSA*
Given data for library development (*Hypothetical reactor name*)	1 BWR (*Titan*) 2 PWRs (*Mimas and Siarnaq*)
Seizure (*Name*)	1 seizure (*Erato*)

Table 2. PCA score of plutonium isotope compositions (Explanatory variable of ^{238}Pu (%), ^{239}Pu (%), ^{240}Pu (%), ^{241}Pu (%), ^{242}Pu (%) were used)



Phase 1 NNFL Creation		**Phase 2** Seizure Analysis
April	May	June
SNF data ✖	Seizure data ✖	Final Report ✖
NNFL creation ⟷ (2-3 weeks)		Seizure analysis ⟷ (2-3 weeks)

The main task of seizure analysis using an NNFL is to determine the consistency of the data between the seized material and the registered materials in the NNFL by comparing patterns or trends in material characteristics. Such patterns or trends can be compared more effectively if key characteristics of each material stored in an NNFL have been identified in advance and added into the database as accompanying information for each material. Although the expertise in the fields of nuclear engineering and chemistry are necessary for seizure analysis, this exercise also taught us that the expertise of those who utilize the NNFL database is equally important, especially regarding the effective utilization and secondary processing of NNFL data. These skills can only be obtained from actual forensics cases or on-the-job training such as that provided in the present exercise. One of the important aspects of NNFL development and its utilization is therefore the accumulation, inheritance, and sharing of such experiences as fingerprint information in traditional forensics.

The purpose of seizure analysis in nuclear forensics is to determine if the seized material is consistent with materials registered in an NNFL. However, as the analysis will exhibit

Table 3. Degree of coincidence for isotope correlations comparison

Grade	Degree of Coincidence
A	90 – 100%
B	70 – 90 %
C	30 – 70 %
D	0 – 30 %

Table 4. Example of isotope correlations comparison

Isotope correlation	BWR (*Titan*)	PWR-1 (*Mimas*)	PWR-2 (*Siarnaq*)
U-235% vs. U236%	D	B	B
Pu/U-238 vs. U-235%	D	B	B
U-235% vs. Pu/U	D	B	C
U-235% vs. Pu-240/Pu-239	D	C	D
U-235% vs. Pu-241/Pu-240	D	D	B
U-235% vs. Pu-241/Pu-240	D	D	B
U-235% vs. Pu-242/Pu-240	D	C	A
U-235% vs. Pu-242/Pu-241	D	C	A

Table 5. Summary of isotope correlations comparison

Result of comparison	No. of items
Stronger consistency with *PWR-1* than *PWR-2*	0
Comparable consistency with *PWR-1* and *PWR-2*	20 (11*)
Stronger consistency with *PWR-2* than *PWR-1*	25
Total	45

* Comparatively stronger consistency with *PWR-2* than *PWR-1*.

uncertainty in most cases, it may be extremely unusual for the specific material to perfectly match the seized material. Therefore, it is necessary to determine the reliability or confidence level for seizure analysis results. An ideal approach is to define a standardized measurement and seizure analysis procedure in order to obtain objective evidence. Although decisions based on the analysis results are subjective to a certain extent, the reliability of the analysis must be validated by showing through quantitative evidence that the results are both reproducible and objective.

The technical lessons learned from the present exercise will be applied to the development of the NNFL at JAEA. A prototype database is currently being developed based on nuclear materials data from fuel cycle facilities at JAEA. As a first step towards NNFL development, a preliminary survey on nuclear materials belonging to the JAEA facilities has been conducted; lessons learned from the exercise were applied to the survey and to the preliminary design of the database. Because the JAEA has a large number of facilities that involve a full-scale nuclear fuel cycle as well as various types of nuclear materials, the prototype database will be large, complex, and highly realistic. The database design (which is still under development) is shown in Figure 3.[7] This database consists of two smaller databases. The first of these databases was named Nuclear Fuel & Fuel Cycle DB and stores raw nuclear materials data and product nuclear fuels in addition to information on their parent processes and parent fuel cycle facilities. The second database was named Sample & Analysis DB, which stores analysis results of nuclear forensics data, quality control, material accounting, and other data sets. The prototype database and its basic associated evaluation system have been completed and will be improved by adding the key characteristic identifier and some development items as follows:

- construction of quantitative measurement for material matching,
- procedures on how to present the results of seizure analysis, and
- implementation of a "knowledge base" for the NNFL system.

A knowledge base is an important item for NNFL improvement and will record operation histories, discussions, and knowledge obtained through seizure analysis in order to provide relevant experiences for subsequent nuclear forensics cases.

The present tabletop exercise was an effective means of learning the overall process, from library creation based on raw nuclear materials data to the seizure analysis for spent nuclear fuels. Furthermore, it is expected that the next exercise of NNFL will be even more effective; as previously mentioned, the task of compiling data into the database was considerably easy because our data set was small and relatively simple in nature. A great deal more experience could be garnered on NNFL database creation if the amount and complexity of the provided data for the hypothetical NNFL creation was increased. Including the various characteristics of isotopes, trace elements, and particles would be a useful improvement in the exercise. It will also be helpful for participants to examine a nation's capability for seizure analysis through the NNFL, as the situation in which the origin of unknown material does not match with NNFL data for a particular nation is a realistic situation of international inquiry. This attempt would help to strengthen international cooperation between members of the nuclear forensics community in the future.

Conclusions

The first virtual, entirely web-based international tabletop exercise on NNFLs, Galaxy Serpent, was held by ITWG and the JAEA participated in the exercise as a part of the NNFL development project in Japan. An NNFL database of provided SNFs on BWR, PWR-1, and PWR-2 was completed in first phase of the exercise. By using isotope correlation comparison technique, in the second phase, it was found that a hypothetical seized nuclear material was definitely consistent with the SNF of PWR-2, a conclusion also confirmed by the PCA method and verified by exercise organizers. The present exercise proved to be an effective means of learning the overall process, from the creation of the NNFL to the seizure analysis for spent nuclear fuels. The JAEA has developed a prototype nuclear material database and several technical lessons learned from this exercise will be applied to the development of the prototype NNFL in the next step of our project. It is expected that the next NNFL exercise will provide a more comprehensive nuclear material data set covering the entire nuclear fuel cycle and will involve an international inquiry system linking the NNFLs for a strengthened implementation of international cooperation in nuclear forensics.

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South Africa's Experiences in the Galaxy Serpent National Nuclear Forensics Library Tabletop Exercise

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Abstract

A South African team participated in the Galaxy Serpent National Nuclear Forensics Library (NNFL) Tabletop Exercise (TTX). Team South Africa, designated as representing the Tucana Galaxy, established an NNFL from supplied data on spent fuel from three fictitious reactors — Iapetus, Hyperion, and Enceladus. Subsequently, Team South Africa was supplied with data from a "seized" sample of spent fuel said to have come from the Erato reactor (pseudonym), and asked to determine if the Erato fuel was or was not consistent with spent fuel data contained in the Tucana NNFL. All the supplied data sets were developed from material in the SFCOMPO website (i.e., public domain data). Comparison of the Erato data with the Tucana NNFL showed a combination of suggestive positive and suggestive negative results. This exercise improved the South Africa Nuclear Forensics team awareness of the need for NNFLs and the methods that can be used to generate one.

Introduction

At ITWG-17, the 2012 annual meeting of the Nuclear Forensics International Technical Working Group (ITWG) held in the Netherlands, South Africa and several other countries volunteered to participate in the Galaxy Serpent, National Nuclear Forensics Library (NNFL) Virtual Tabletop Exercise (TTX). The TTX was conducted under the auspices of the ITWG and co-led by the U.S. State Department and the U.S. Department of Homeland Security. Teams of scientists from nine of the countries/nations, along with one regional institute, that volunteered completed the exercise and agreed to contribute papers on their experiences.

Galaxy Serpent participants established a model NNFL from three data sets provided to them by the exercise organizers.[1] The data sets were from spent fuel from three hypothetical reactors in their "galaxy." Participants were then given a fourth data set, said to be from a "seized" sample, and asked to determine if it was or was not consistent with the spent fuel data in their NNFL. The data sets organizers supplied to the participants originated from the SFCOMPO public domain website. The data sets were altered and re-purposed for this nuclear forensics application before being supplied to participants.[1]

Team South Africa developed the Tucana Galaxy NNFL from supplied data on the so-called Iapetus, Hyperion, and Enceladus reactors. These spent fuel power reactors data collection included data from two boiling water reactors (BWR), Enceladus and Hyperion, and data from one pressurized water reactor (PWR), Iapetus. The "seized" sample, which was compared with the NNFL was designated as coming from the Erato reactor.

Team South Africa took the position of observer during the first round of the Galaxy Serpent TTX, and participated as a direct participant team in the second round starting in April 2013. The first four weeks, designated phase 1, involved working with provided data sets and developing a model NNFL. In phase 2, data for a simulated border seizure of spent nuclear fuel material were posted to the South Africa galaxy's workspace at the start of the fifth week for analysis and final reporting of the findings. Four weeks was given for the task of determining whether data for the hypothetical seizure was or was not consistent with reactors in the Tucana NNFL.

Method of Data Analysis

Team South Africa successfully developed the Tucana Galaxy NNFL by preparing spreadsheets and graphical plots of data from the three assigned spent fuel power reactors, Iapetus, Hyperion, and Enceladus. These were to be used to interrogate and subsequently determine if the hypothetical seized spent fuel material from a pseudo reactor code named Erato for the purpose of the TTX was or was not consistent with material from Iapetus, Hyperion, and/or Enceladus as captured in the Tucana NNFL. Team South Africa used graphical correlation of isotopic parameters for comparing seized material isotopic data with iso-

Table 1. A representative sample of the tabulated data format that was used to facilitate the isotopic parameter correlations

Hyperion Reactor Data					
^{235}Upercent	^{236}Upercent	burnup (^{148}Nd)	^{235}U/^{238}U	^{236}U/^{238}U	^{239}Pu/Utotal
0.6757	0.3389	27.4	0.0068	0.0034	0.555
0.6273	0.3375	25.7	0.0063	0.0034	0.556
0.9825	0.3044	21.2	0.01	0.0031	0.641
0.9019	0.3099	22.3	0.0091	0.0031	0.624
0.8625	0.3257	23	0.0087	0.0033	0.607
0.8687	0.3237	23.5	0.0088	0.0033	0.615
0.6663	0.3368	25.2	0.0067	0.0034	0.57
0.9877	0.2973	19.9	0.01	0.003	0.636
0.8602	0.3145	20.3	0.0087	0.0032	0.614
1.319	0.319	14.4	0.0134	0.0025	0.721
1.2487	0.2559	15.8	0.0127	0.0026	0.699
1.0359	0.2779	17.5	0.0105	0.0028	0.652

Hyperion Reactor Data (continued)						
^{239}Pu/Pu total	^{239}Pu/^{238}U	^{242}Pu/^{239}Pu	^{240}Pu/^{239}Pu	^{241}Pu/^{240}Pu	^{242}Pu/^{241}Pu	^{137}Cs/U total
0.5556	0.005	0.0938	0.4521	0.5253	0.3947	0.0009
0.5557	0.0039	0.0885	0.4866	0.4352	0.4179	0.0011
0.6413	0.0056	0.4124	0.3446	0.4743	0.2523	0.0008
0.6241	0.0053	0.0469	0.3698	0.4677	0.2713	0.0008
0.6071	0.0052	0.0544	0.3996	0.4611	0.3011	0.001
0.6148	0.005	0.051	0.3838	0.4622	0.2877	0.0009
0.5696	0.0048	0.0742	0.4648	0.4313	0.3708	0.001
0.636	0.0046	0.0455	0.3462	0.5007	0.2624	0.0008
0.6138	0.0038	0.05	0.4038	0.4163	0.2974	0.0008
0.7211	0.0049	0.019	0.2479	0.4681	0.1639	0.0006
0.6992	0.0046	0.0225	0.2793	0.4387	0.1838	0.0007
0.6516	0.0043	0.0369	0.3453	0.4194	0.255	0.0007

topic data already available in the completed Tucana NNFL.

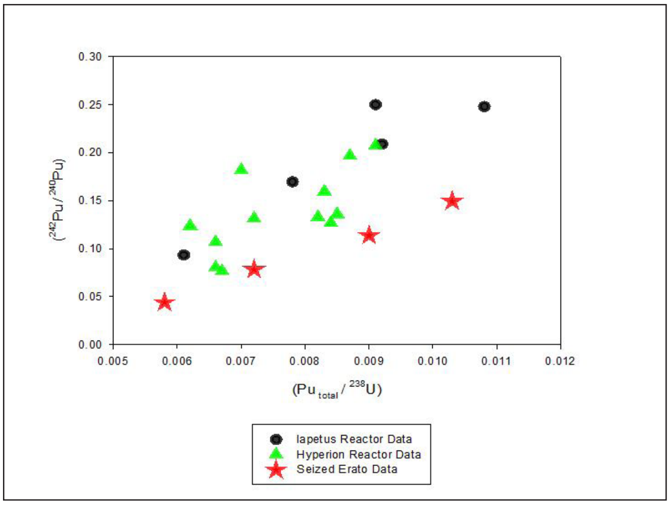
Investigation of the seized material from Erato reactor was based on the significant fissile isotopic parameters correlation understood to allow discrimination amongst data of the same spent fuel origin.[2,3] These include, but not limited to, the following uranium isotope/total uranium and/or Pu isotopes ratios that have also been suggested for use at the ITWG–17 meeting in October 2013:[1,4]

^{235}U/Utotal, ^{234}U/Utotal, ^{236}U/Utotal, ^{234}U/^{238}U, ^{235}U/^{238}U, ^{236}U/^{238}U ^{240}Pu/Putotal, ^{241}Pu/Putotal, (Pu/U) total, ^{242}Pu/^{241}Pu, ^{242}Pu/^{240}Pu, ^{238}Pu/Pu total

The defined approach required that the spent fuel analytical data presented in SFCOMPO first be assessed and organized in a way that is suitable for comparative analysis using key isotopic variables and parameters. The organization of the supplied data included:

- identifying significant isotopic parameters reported for at least two of the reactor of interest to facilitates correlation,
- rearranging the data sets into Excel to facilitate multiple plots of the Tucana Galaxy spent fuel isotopic parameters

Figure 1. Plot of (^{242}Pu/^{240}Pu) against (Pu$_{total}$/^{238}U)



- on the same set of axes (cf. Table 1 for typical layout),
- plotting the data to yield the correlations that are used for analysis of the seizure material.

The data were sorted on a "per sample" basis by assigning a group of parameters to a single sample. The cooling down information provided some level of validation that each of the parameters being assigned to the same sample came from the same power reactor spent fuel.[5] Figure 1 shows illustrative data from the Iapetus reactor after these were rearranged to meet the established Excel spreadsheet format for data analysis. Note that Table 1 provides a representative sample and not a complete data set.

Findings of the Hypothetical Source Seizure Analysis

Figures 1 through 12 show the isotopic ratio patterns of the Tucana NNFL and the seized Erato sample. These were aimed at seeking possible consistency of the isotopic parameters of the intercepted hypothetical spent fuel from Erato reactor.[1] Comparative analysis of isotopic parameter correlations with each of the three spent fuel reactor data collected in the Tucana NNFL yielded a number of observations, insights, and conclusions (refer also to Table 2 for a summary of the observations). In the figures and discussions which follow, confidence levels were assigned using an ITWG establish convention based on a five-point scale.[6]

Examination of the plot of (^{242}Pu/^{240}Pu) against (Pu total/^{238}U) shown in Figure 1 shows a suggestive positive that the Erato spent fuel seized material could have come from the Hyperion reactor or the Iapetus reactor. The Enceladus reactor is not included because these isotopic ratios were not available from the SFCOMPO website.

Figure 2. Plot of ^{235}U against (^{242}Pu/^{241}Pu)

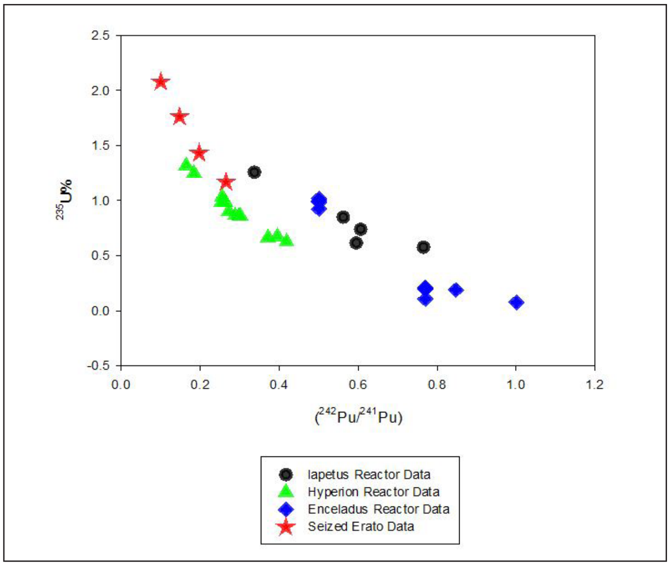
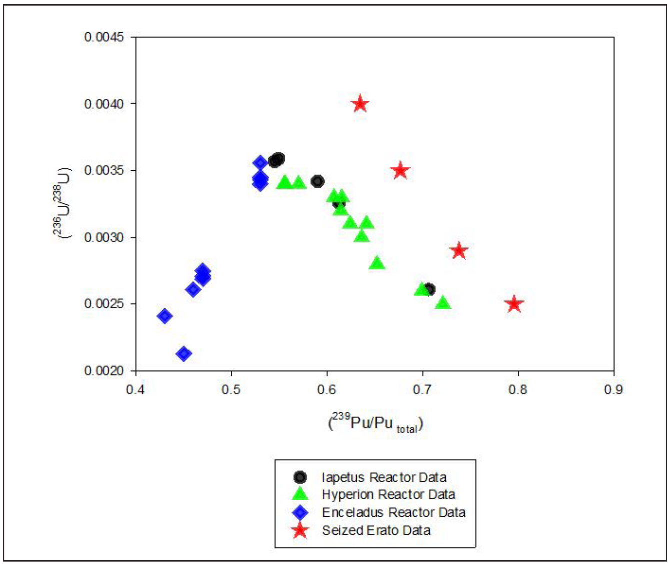


Figure 3. Plot of (^{236}U/^{238}U) against (^{239}Pu/Pu$_{total}$)



The correlation plot of ^{235}Upercent against (^{242}Pu/^{241}Pu) shown in Figure 2 shows a conclusive negative that the seized material originated from the Enceladus reactor. The plot shows a suggestive positive chance of the material coming from the Hyperion than from the Iapetus reactor.

The plot of (^{236}U/^{238}U) against (^{239}Pu/Pu total) shown in Figure 3 shows a conclusive negative for the Enceladus reactor being the possible origin of the spent Erato fuel. The plots could also not discriminate between the Hyperion and the Iapetus reactor since the fittings are inconclusive.

Table 2. Summary of the isotopic parameters correlation findings of the Tucana Galaxy

Plot number	ERATO data relationship to		
	Iapetus data	**Hyperion data**	**Enceladus data**
1	suggestive positive	suggestive positive	–
2	inconclusive	suggestive positive	conclusive negative
3	inconclusive	inconclusive	conclusive negative
4	suggestive positive	conclusive negative	–
5	suggestive negative	suggestive negative	–
6	suggestive positive	conclusive negative	–
7	suggestive positive	suggestive positive	conclusive negative
8	inconclusive	Inconclusive	Inconclusive
9	suggestive negative	suggestive positive	suggestive negative
10	inconclusive	Inconclusive	Inconclusive
11	suggestive positive	suggestive positive	–
12	suggestive positive	conclusive negative	–

Plots of ^{235}U percent against (^{239}Pu/U total) in Figure 4 support the suggestion that the spent Erato fuel material comes from the Iapetus, and not the Hyperion reactor. The percent ^{235}U Erato data are of concern in that they are positioned within the ranges from 1.1 to about 2.1; and these are higher than the values reported for the Iapetus with only one entry covered in the upper percent ^{235}U range — hence the match is only suggestive positive for Iapetus and conclusive negative for Hyperion as the possible origins of the Erato fuel material.

The correlation based on the plots of percent ^{235}U against (Pu total/^{238}U) shown in Figure 5 are suggestive negative that the Erato material came from the Iapetus or the Hyperion reactors.

Figure 6 shows the plot of percent ^{236}U against (^{239}Pu/Utotal) is suggestive positive that the Erato material may have originated from the Iapetus reactor as opposed to the Hyperion reactor. Again, no corresponding isotopic ratios were available for the Enceladus reactor from the SFCOMPO website.

The plot of ($^{242}Pu/^{240}Pu$) against burnup shown in Figure 7 is conclusive negative that the seized Erato material originated from the Enceladus reactor. It is however difficult to distinguish the goodness of the suggestive positive fit of the data to the Iapetus and the Hyperion reactors.

Correlation of the plots of ($^{236}U/^{238}U$) against the burnup given in Figure 8 suggests that the Erato material has an inconclusive chance of coming from any one of the three reactors in the national nuclear forensics library (i.e., the Iapetus, Hyperion and Enceladus reactors database).

Figure 4. Plot of % ^{235}U against ($^{239}Pu/U_{total}$)

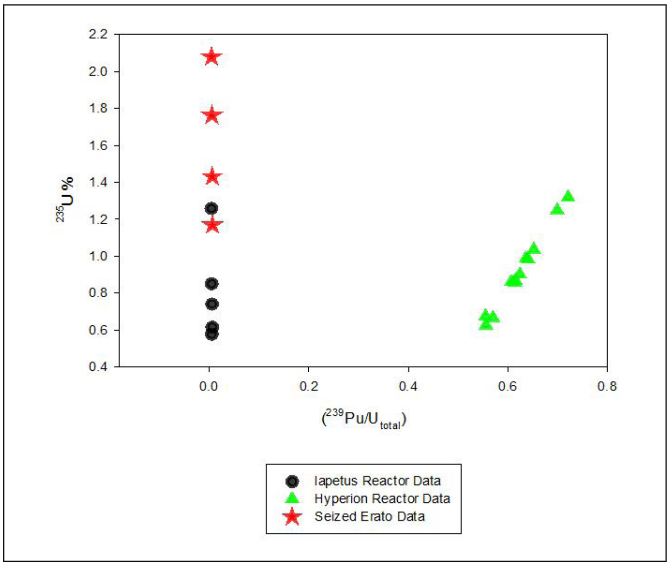
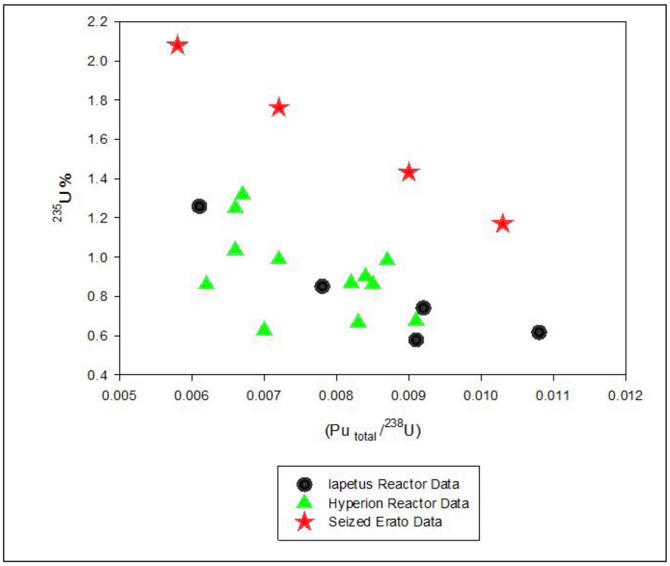


Figure 5. Plot of % ^{235}U against ($Pu_{total}/^{238}U$)



The plots of (^{239}Pu/Pu total) against burnup that are shown in Figure 9 are suggestive positive that the Erato material originated from the Hyperion than it could be from the Iapetus or the Enceladus reactors.

Plots of percent ^{235}U against burnup shown in Figure 10 suggest that the plotted parameters are not suitable for use in distinguishing between the origins of the seized fuel materials.

The correlation based on the plots of (Pu total/^{238}U) against burnup was also not useful in resolving the situation as there is suggestive positive for both reactors in Figure 11.

Figure 6. Plot of %^{236}U against (^{239}Pu/U$_{total}$)

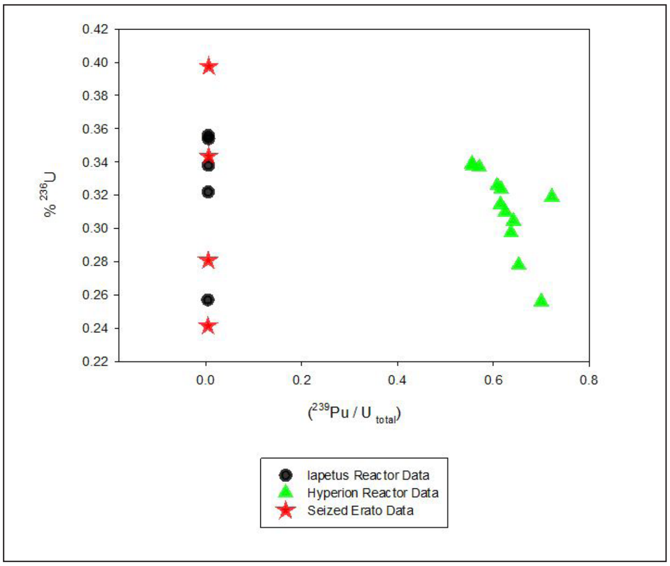


Figure 7. Plot of (^{242}Pu/^{240}Pu) against burnup (^{148}Nd)

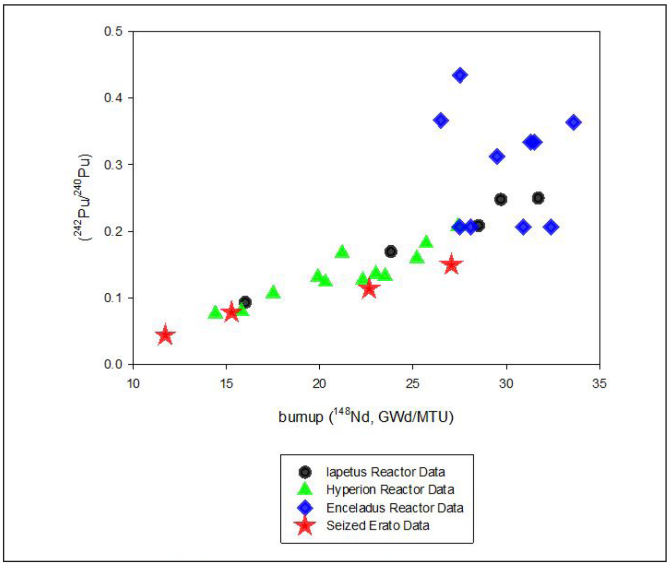


Figure 8. Plot of (^{236}U/238) against burnup (^{148}Nd GWd/MTU)

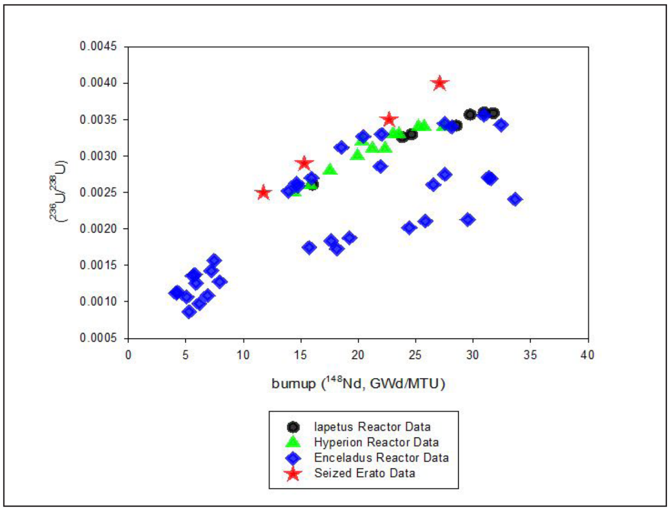
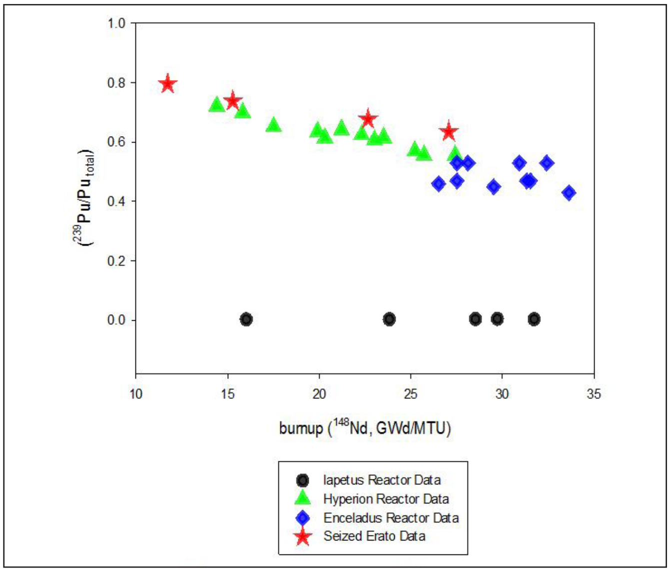


Figure 9. Plot of (^{239}Pu/Pu$_{total}$) against burnup (^{148}Nd GWd/MTU)



Correlation based on plotting (^{239}Pu/U total) against burnup shown in Figure 12 is suggestive positive that the seized Erato material originated from the Iapetus and not the Hyperion reactor (conclusive negative). This is much in agreement with the findings shown in Figure 6.

The correlations based on the isotopic parameters provided inadequate information for our use in deducing if the seized material shares a common origin with the material in our NNFL. This motivated us to investigate the correlation of the isotopic ratios of fissile isotopes such as ^{239}Pu, ^{240}Pu, ^{236}U, and ^{235}U against the burnup (as determined by the ^{148}Nd analysis method). Results of the analysis of the plots of the isotopic parameters in Figures 1 through 12 are summarized in Table 2.

The Rationale of Our Assessment Findings and Concluding Remarks

Whilst the approach followed can tolerate serious data gaps between seizure data and the NNFL database, the correlation plots shown in Figures 1 through Figure 12, when considered collectively, suggest that it is not possible to determine the origin of the seized fuel material based solely on the isotopic parameters reported in the NNFL developed for the TTX. Attempts to make use of different isotopic parameters in isolation generally led to contradicting results. For instance, the

Figure 10. Plot of ^{235}U % against burnup (^{148}Nd GWd/MTU)

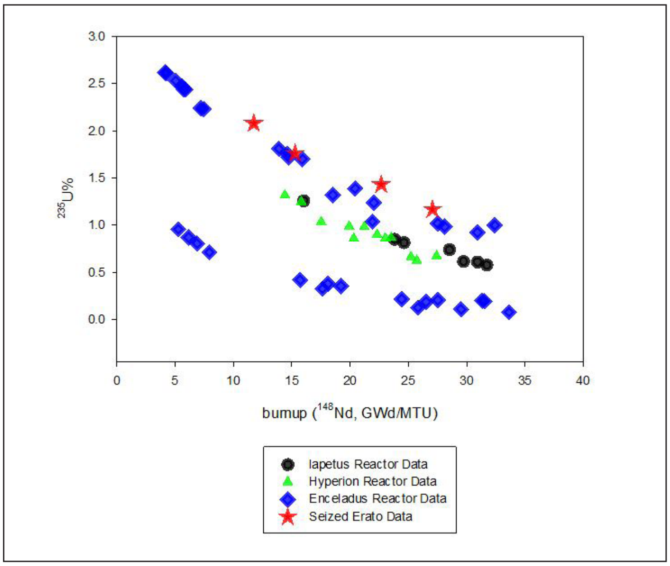


Figure 11. Plot of (Pu_{total}/^{238}U) against burnup (^{148}Nd GWd/MTU)

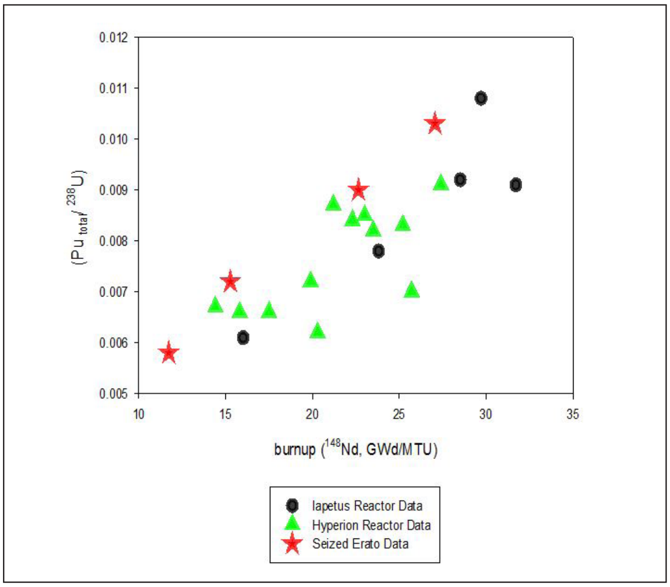
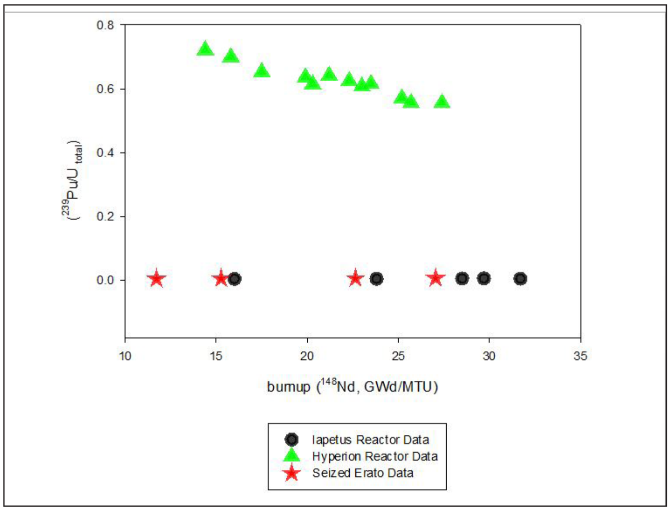


Figure 12. Plot of (^{239}Pu/Utotal) against burnup (^{148}Nd GWd/MTU)



results plotted in Figures 6 and 12 show suggestive positive for Iapetus as the origin of the seizure when compared to the conclusive negative for the Hyperion reactor. This observation contradicts the findings shown in Figure 9, which points at Hyperion as the suggestive positive origin of the hypothetical seized material versus suggestive negative for the Iapetus and the Enceladus reactors.

Furthermore, in case of the Hyperion reactor the isotopic data plotted in Figures 4 and 5 suggested negative correlation as most of the characteristics disagree. The isotopic parameters plotted in Figures 6, 7, 11, and 12 agreed with the proposition of the Iapetus reactor as the likely origin of the hypothetical seizure. For the Enceladus reactor, correlation plots (refer to Figures 2, 3, and 7) results are conclusively negative as most of the considered isotopic correlations for the seized spent fuel material disagreed with those in the NNFL. The correlations shown in Figures 1, 7, and 11 are suggestive positive for both the Iapetus and the Hyperion reactors as most of the isotopic parameters matched those of the seized Erato spent fuel. Meanwhile, both the Hyperion and the Iapetus reactors isotopic parameters plotted in Figure 5 show suggestive negative match with the Erato isotopic parameters.

All the analysis approaches used (i.e., isotopic correlation and isotopic ratios versus burnup) led to the same conclusion that the Erato (seized spent fuel) is not consistent (conclusive negative) with the parameters for the Enceladus reactor spent fuel, but closely resembled spent fuel isotopic parameters for the Iapetus and the Hyperion reactors. Findings of the correlation of isotopic behavior of Erato to the NNFL can be summarized as follows:

Fitting of isotopic parameters	poorest for Enceladus	conclusive negative
	somewhat consistent with Hyperion	suggestive negative
	best fit to the Iapetus but with conflicting behavior	suggestive positive

The seized Erato spent fuel is suggestively positive to have come from the Iapetus reactor, suggestively negative to have come from Hyperion, and conclusively negative to have come from the Enceladus reactor. In other words, data analysed based on the above-named isotopic parameters graphical correlations conclusively eliminate the Enceladus reactor as the origin of the seized Erato spent fuel material. These findings, combined with reactor-type information noted above, namely that Enceladus and Hyperion are BWRs, while Iapetus is a PWR, gives important insights. Specifically, isotopic correlations, considered as a group, strongly suggest that the Erato seizure did not come from a BWR source, and is consistent with a PWR. The solution for this round, as given by exercise organizers,[4] was that the seizure did not originate from a reactor in the Tucana galaxy, but did originate from a PWR assigned to another galaxy in this round. While our findings were not definitive, they are certainly consistent with the revealed solution.

It follows that the origin of the seized material could not be linked to the Tucana NNFL. These findings can be attributed to inadequate information available about the analytical methods used to gather the open literature isotopic parameters in the provided data sets and perhaps also to the unsuitability of some of the correlations chosen for this comparative analysis. There are also certain pieces of data that showed remarkable deviation from the rest of the data related to a particular sample. In a real library investigation, inadequate, in terms of library holdings, or incomplete, in terms of data for a given sample, would normally be easy to correct. Further investigations could also focus on morphological information, determination of elemental impurities, trace elements analysis, microscopic studies, etc. However, such information would be far more valuable and useful if collected and integrated prior to an actual nuclear security event.

Lessons Learned from the TTX

The Galaxy Serpent TTX served well as a valuable tool to facilitate an improved understanding of the NNFL development process and the interrogation of the library pertaining seized material analysis. Spent fuel data can be organized into a useful NNFL to support seized spent fuel data query and related analysis.

Building the initial NNFL required good insight into the characteristic features that have the potential to distinguish between nuclear materials of different origin, such as the significant fissile isotopic parameters discussed earlier. Use of an existing NNFL is not easy as it require special knowledge of spent fuel nuclear material chemistry. Knowledge of the different analytical techniques used to characterize the spent fuel materials is also deemed helpful. In the case of South Africa, it was found to be particularly important to first prioritize the relevant data type from the large amount of provided information.[2] It was also necessary to first reorganize the SFCOMPO data for a nuclear forensics application to facilitate the TTX development. The prioritized data had to be processed to provide for easy NNFL development and for further processing such as graphical correlations.

Subject matter experts (SME) are therefore also required to help ascribe uniform uncertainty values in data reported in the SFCOMPO public domain database, which may not be consistent as it was gathered for fuel post irradiation studies, and repurposed here for a nuclear forensics application. The "limited universe" of data, composed of only low-enriched uranium-based reactors, is best interpreted by scientists with specific expertise. Thus, an expanded pool of experts, such as fuel engineers and statisticians, could provide additional insights in advancing a more diverse NNFL in this domain.

While this exercise employed a limited, model NNFL, in terms of the number of reactors used and data associated with each, and was conducted over a two-month period, some important conclusions were reached. Namely, the small size of the database comprising the NNFL led to inconsistent results for the hypothetical seizure when considering individual isotopic correlations. When isotopic correlations were considered in total, while they did not provide certainty, certain trends did emerge, as noted earlier. Further, when the comparative trends of all reactors relative to the seizure were considered, it was highly suggestive that the seizure did not originate from a BWR, and likely originated from a PWR. As noted, the findings of the South African team were consistent with the solution provided by exercise organizers.

The tabletop exercise raised awareness about the technical aspects of creating and using NNFLs including the challenges associated with the use of limited data sets of spent nuclear fuel data, but also illustrated that a rudimentary NNFL is preferable to not having one. The TTX also expanded the community of nuclear forensics awareness and expertise by providing opportunities for participation by junior members and non-forensics personnel who otherwise would not have been involved in nuclear forensics capacity building efforts.

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The UK's Contribution to the Galaxy Serpent Tabletop Exercise

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Abstract

This paper summarizes the United Kingdom's (UK) contribution to the Galaxy Serpent Virtual Tabletop Exercise. The UK team consisted of representatives from the Atomic Weapons Establishment (AWE), National Nuclear Laboratory (NNL), and the University of Liverpool. Each institution in the UK team approached the Galaxy Serpent exercise independently with a number of different methodologies: Radiochemistry and Isotopic Correlations (AWE), Reactor Type Multivariate Analysis (NNL), and Laplacian Eigenmaps (University of Liverpool). Despite the variety of methodologies employed, all three methodologies provided data that supported the same conclusion, namely that the seized fuel pin correlated at a suggestive positive level with a particular reactor. Other supporting sources of reactor data, although available to the UK team, were not fully explored. As a full exercise data set was not provided to the UK, i.e., the seized fuel pin was not in the UK data set, it was not possible for the UK to conclude that the seized sample was definitely in the UK data set. This paper details the methodologies employed by each institution, the UK team's decision process, and an explanation of the misleading conclusion.

Introduction

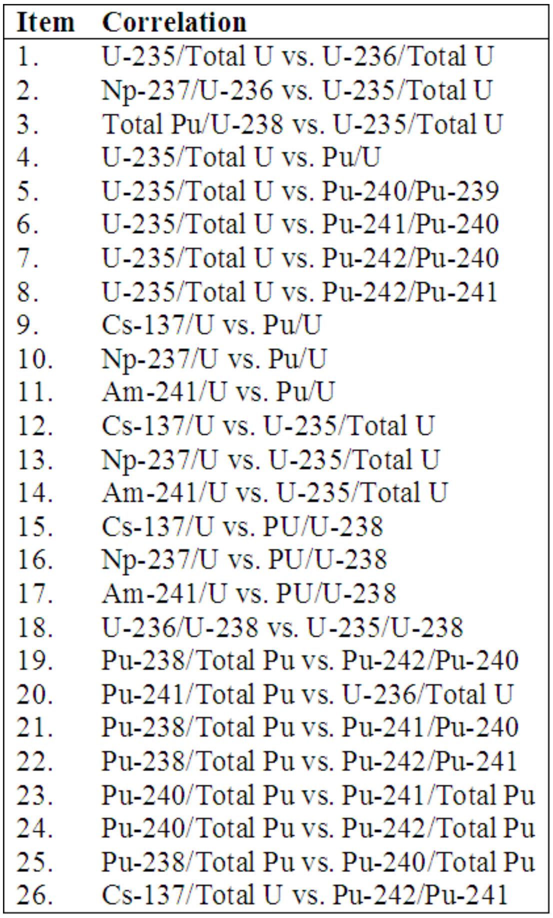
The UK team conducted the exercise between April and June 2013. The UK team was split into three groups and composed of representatives from the Atomic Weapons Establishment (AWE), National Nuclear Laboratory (NNL), and the University of Liverpool. A full data set was not provided to the UK. Instead, data from three reactors were provided. The reactors allocated to the UK in Galaxy Serpent were: Anthe (pressurized water reactor (PWR)), Tethys (PWR), and, Janus (boiling water reactor (BWR)). Data from these reactors were supplied as Microsoft Excel files, and typically included actinide isotopic compositions, selected fission product quantities, and burnup measurements. Comparisons were then made with a hypothetically seized fuel pin (Erato) that had been isotopically characterized at four positions in order to establish whether the seized material was consistent with any of the reactors for which the UK had been given data. In order to achieve this, each institution in the UK team worked independently with different approaches — radiochemistry and isotopic correlations (AWE), reactor type multivariate analysis (NNL), and Laplacian Eigenmaps (University of Liverpool). A discussion of each of these methodologies follows.

Radiochemistry and Isotopic Correlations

The reactor data, including the Erato data when available, were processed in a Microsoft 2003 Excel Workbook using lookups, advanced filters, and Visual Basic for Applications (VBA) code. The isotopic correlations relating to fluence, reactor type, or fuel enrichment detailed in Table 1 were plotted. It was observed that the Tethys reactor data could be subdivided into two populations for many of these plots with the first population consisting of Tethys samples 1-5 and 11-16, and the second population consisting of Tethys samples 6-10. Isotopic correlations were plotted using the Tethys data split into two populations. The usefulness of selected correlations will now be discussed.

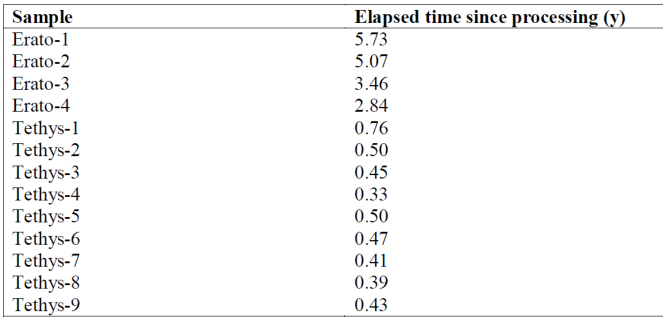
Figure 1 is a plot of Pu-238/Total Pu vs. Pu-242/Pu-240. According to Wallenius Peerani and Koch,[1] this plot can discern reactor type, however this is aimed mainly at distinguishing gas-cooled/graphite-moderated reactors from water cooled and moderated reactors and poorly resolves BWR and PWR reactors. The Erato data are plotted along with the three refer-

Table 1. The Isotopic correlations examined



Item	Correlation
1.	U-235/Total U vs. U-236/Total U
2.	Np-237/U-236 vs. U-235/Total U
3.	Total Pu/U-238 vs. U-235/Total U
4.	U-235/Total U vs. Pu/U
5.	U-235/Total U vs. Pu-240/Pu-239
6.	U-235/Total U vs. Pu-241/Pu-240
7.	U-235/Total U vs. Pu-242/Pu-240
8.	U-235/Total U vs. Pu-242/Pu-241
9.	Cs-137/U vs. Pu/U
10.	Np-237/U vs. Pu/U
11.	Am-241/U vs. Pu/U
12.	Cs-137/U vs. U-235/Total U
13.	Np-237/U vs. U-235/Total U
14.	Am-241/U vs. U-235/Total U
15.	Cs-137/U vs. PU/U-238
16.	Np-237/U vs. PU/U-238
17.	Am-241/U vs. PU/U-238
18.	U-236/U-238 vs. U-235/U-238
19.	Pu-238/Total Pu vs. Pu-242/Pu-240
20.	Pu-241/Total Pu vs. U-236/Total U
21.	Pu-238/Total Pu vs. Pu-241/Pu-240
22.	Pu-238/Total Pu vs. Pu-242/Pu-241
23.	Pu-240/Total Pu vs. Pu-241/Total Pu
24.	Pu-240/Total Pu vs. Pu-242/Total Pu
25.	Pu-238/Total Pu vs. Pu-240/Total Pu
26.	Cs-137/Total U vs. Pu-242/Pu-241

Table 2. Elapsed time since last processing from Am-241 and Pu-241



Sample	Elapsed time since processing (y)
Erato-1	5.73
Erato-2	5.07
Erato-3	3.46
Erato-4	2.84
Tethys-1	0.76
Tethys-2	0.50
Tethys-3	0.45
Tethys-4	0.33
Tethys-5	0.50
Tethys-6	0.47
Tethys-7	0.41
Tethys-8	0.39
Tethys-9	0.43

ence reactors data in Figure 1. From this plot Janus data could be separated from the remaining reactor data due to the Janus data set being closer to the origin, however this is a function of length of irradiation of that sample rather than the isotopics of the reactor type.

From the plot of U-235/Total U vs. Total Pu/Total U (Figure 2), it is evident that the Janus data set possesses a lower Total Pu/Total U range of values than any of the PWR reactors and Erato. Similarly, plots of Pu-241/Total Pu vs. U-236/Total U (Figure 3) and U-235/Total U vs. Pu-241/Pu-240 (Figure 4) clearly distinguish between Janus and the other reactor data, including Erato. For all three plots, Erato remains close to the PWR data, specifically that of the Tethys reactor. The similarity of the Tethys and Erato data in plots of Pu-238/Total Pu vs. Pu-241/Pu-240, and Pu-238/Total Pu vs. Pu-242/Pu-241 was taken as showing a correlation between the two reactors (Figures 5 and 6).

Bi-variate plots with Pu-239, Pu-240, Pu-241 and Pu-242 distinguish between the reactor materials and reduce the influence of initial fuel enrichments on the isotopic correlations. Figure 7 is a plot of Pu-240/Total Pu vs. Pu-241/Total Pu. The plots show the three reactors (Anthe, Janus and Tethys) are clearly resolved with the seized sample, Erato, overlapping the Tethys reactor data. Decay correction of the Pu data in Figure 7 with the cooling times of the reactors and an NNL estimated cooling time of two years for the seized fuel pin did not alter the observed correlation between Tethys and Erato.

Other plots were less useful due to either the lack of data for the BWR reactor, Janus, or the poor discrimination between the reactors and Erato. Examples of this include U-235/Total U vs. Pu-242/Pu-241 (Figure 8) where there is overlap between Erato and all of the reactors and Cs-137/Total U vs. Total Pu/Total U where although there is reasonable distinction between Tethys and Erato, no data are available for Janus (Figure 9).

Discrepancies arising from other isotopic correlations are challenging to explain. A plot of Am-241/Total U vs. U-235/Total U (Figure 10) shows that the Erato data no longer correlate well with the Tethys reactor. The source of this discrepancy is not obvious but it is possible that the poor correlation in this plot is due to a combination of irradiation history and post irradiation cooling time or sample age (via a plot of Pu-238/Total Pu vs. Am-241/Pu-241, Figure 11). The time in years elapsed from last processing derived from the Am-241 and Pu-241 data for each Erato and Tethys measurement is given in Table 2. Except for Am-241 and Pu-241, most chronometers are not valid as the plutonium daughters are all uranium isotopes. Decay correction of the data using the cooling times of the reactors and an NNL estimated cooling time of two years for the seized fuel pin did not improve the correlation between Tethys and Erato.

A plot of U-236/U-238 vs. U-235/U-238 reveals that the Tethys data are split into two populations bordering the Erato data (Figure 12). Assuming negligible U-236 is present in the

Figure 1. Pu-238/Total Pu vs. Pu-242/Pu-240 for the reactors and the seized fuel pin

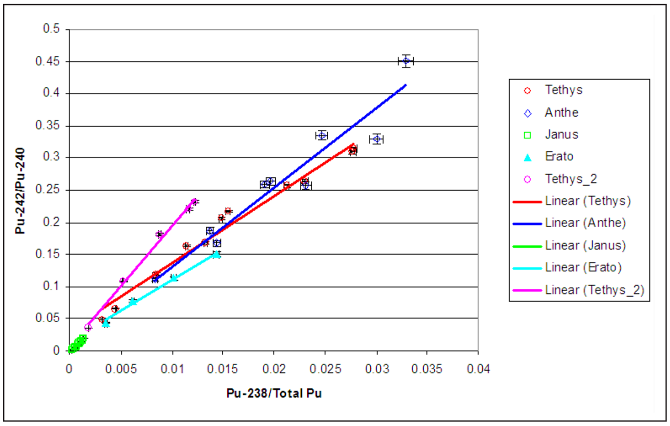


Figure 2. U-235/Total U vs. Total Pu/Total U for the reactors and the seized fuel pin

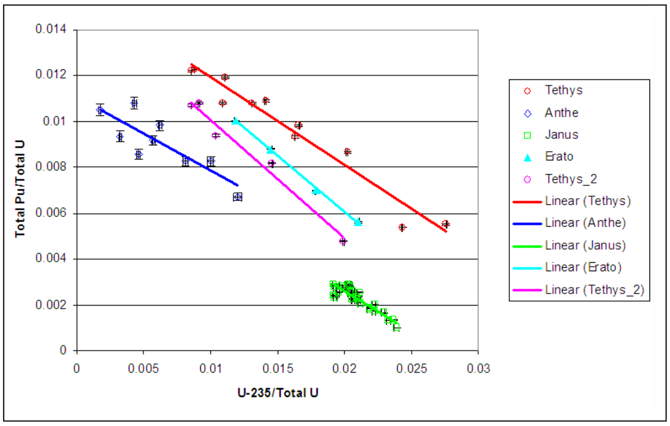


Figure 3. Pu-241/Total Pu vs. U-236/Total U for the reactors and the seized fuel pin

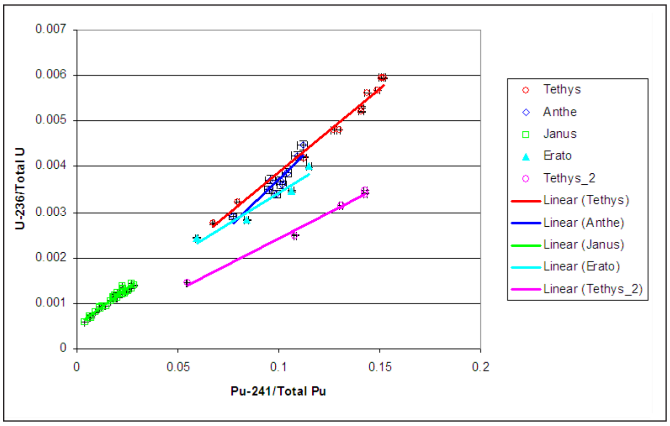


Figure 4. U-235/Total U vs. Pu-241/Pu-240 for the reactors and the seized fuel pin

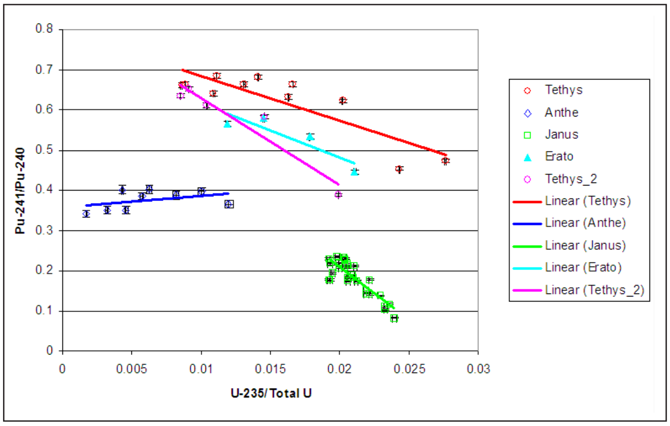


Figure 5. Pu-238/Total Pu vs. Pu-241/Pu-240 for the reactors and the seized fuel pin

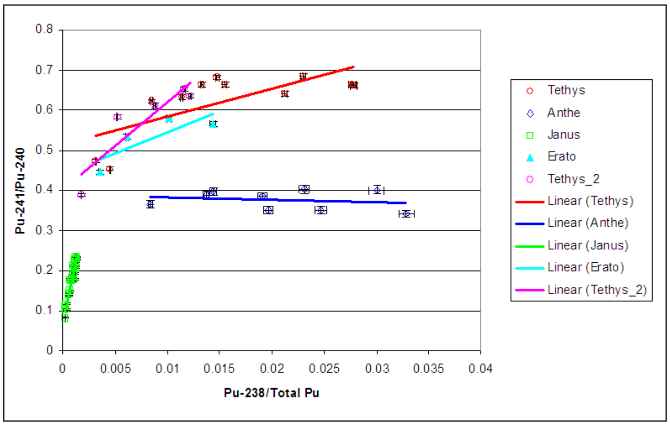
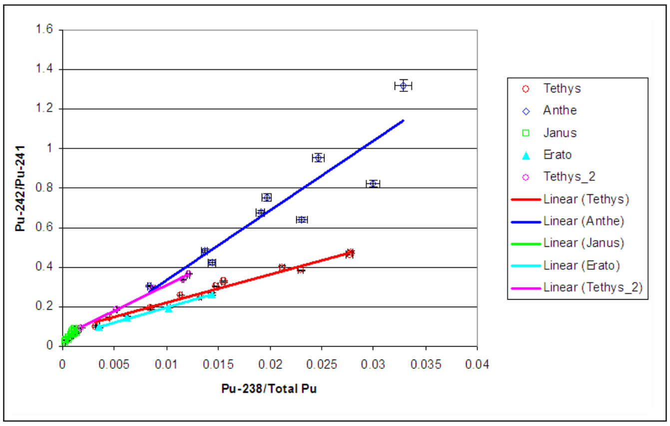


Figure 6. Pu-238/Total Pu vs. Pu-242/Pu-241 for the reactors and the seized fuel pin



fuel prior to irradiation; the intercept on the ordinate axis is a measure of initial U-235 fuel enrichment. The two Tethys populations indicate an enrichment of 2.9 and 4.6 percent. From this plot, Erato has an enrichment of 3.5 percent. The initial enrichment of Erato is between the two populations of Tethys. If the operating records for the Tethys reactor were available they could be examined to establish whether any fuel with an enrichment of 3.5 percent had been used in the reactor, and if

Figure 7. Pu-240/Total Pu vs. Pu-241/Total Pu for the reactors and the seized fuel pin

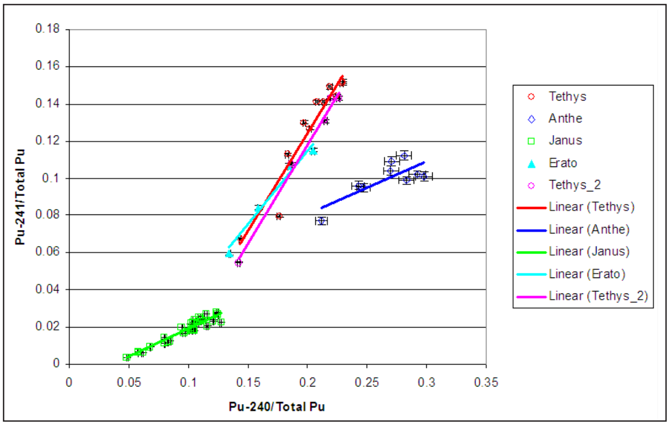


Figure 8. U-235/Total U vs. Pu-242/Pu-241 for the reactors and the seized fuel pin

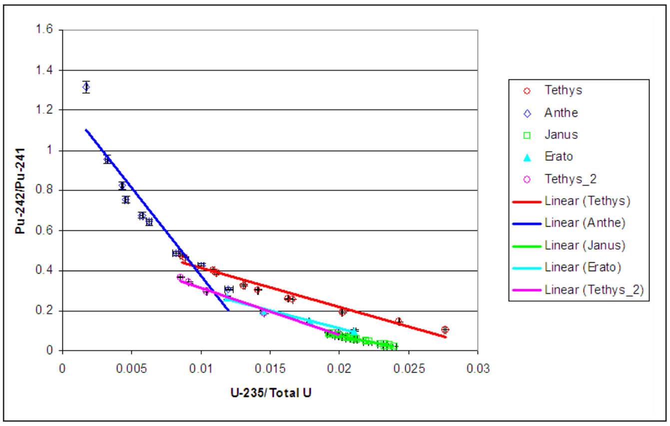


Figure 9. Cs-137/Total U vs. Pu-242/Pu-241 for the reactors and the seized fuel pin

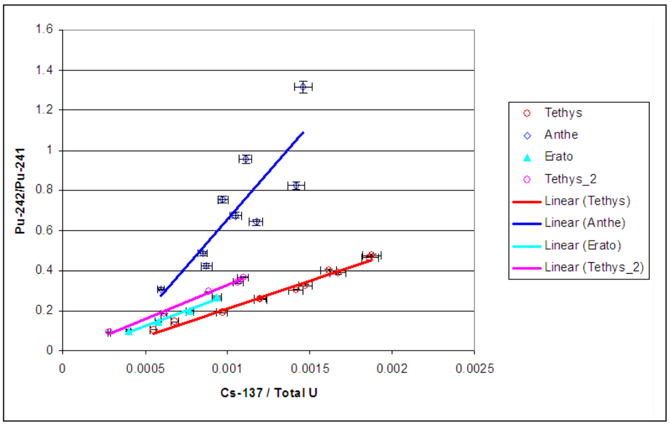


Figure 10. Am-241/Total U vs. U-235/Total U for the reactors and the seized fuel pin

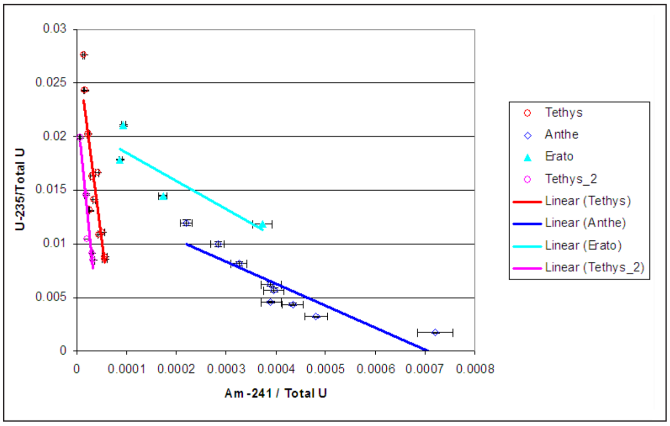


Figure 11. Pu-238/Total Pu vs. Am-241/Pu-241 plots for the reactors and the seized fuel pin

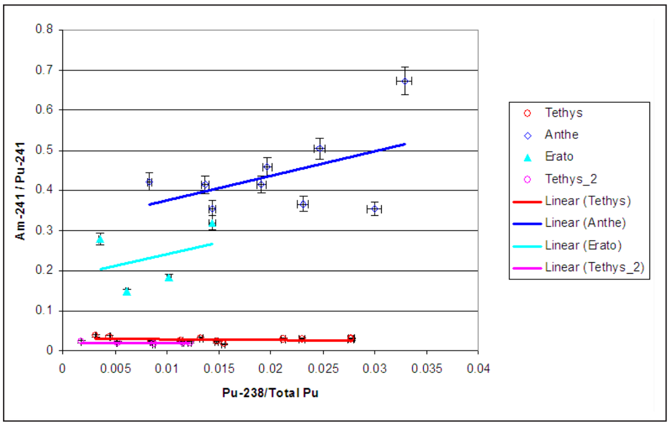
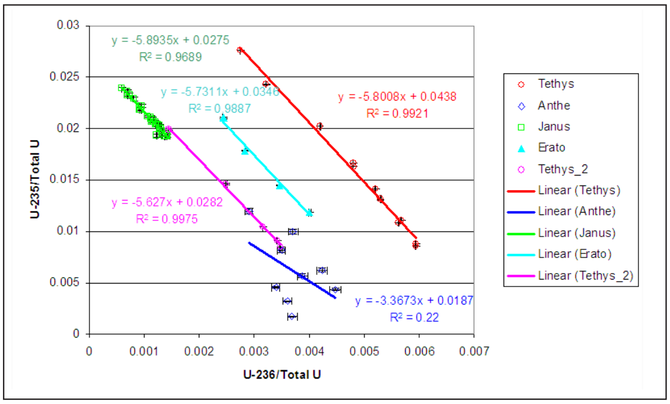


Figure 12. U-236/Total U vs. U-235/Total U for the reactors and the seized fuel pin. The colour of each regression equation matches its equivalent regression line.



the differing enrichments of the Tethys data and the seized fuel pin Erato provide a suitable means of discrimination.

The isotopic correlations examined allowed the elimination of the Janus reactor data as these were observed to be least correlated to Erato. With Janus eliminated, linear discriminant analysis (LDA)[2] was applied to selected data sets to establish whether the Erato data were consistent with either the Tethys or the Anthe reactors.

Figure 13. U-235/Total U vs. Total Pu/Total U LDA plot

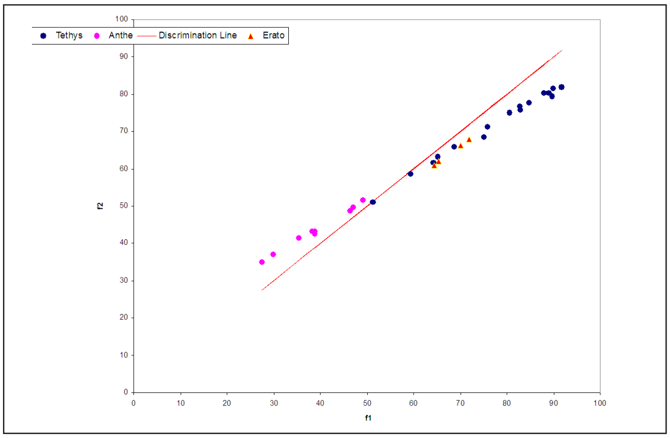


Figure 14. U-235/Total U vs. Pu-241/Pu-240 LDA plot

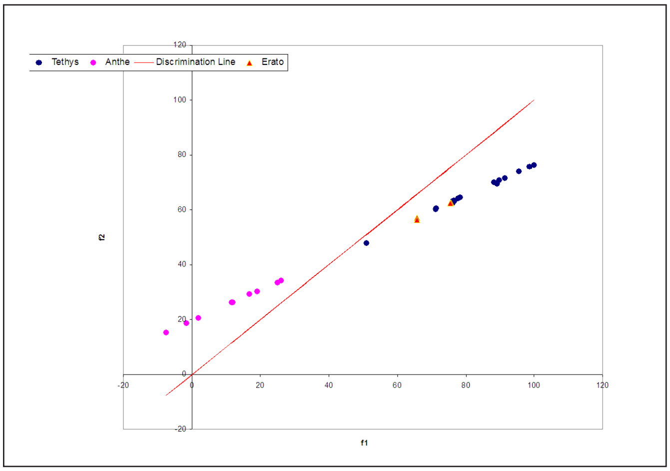
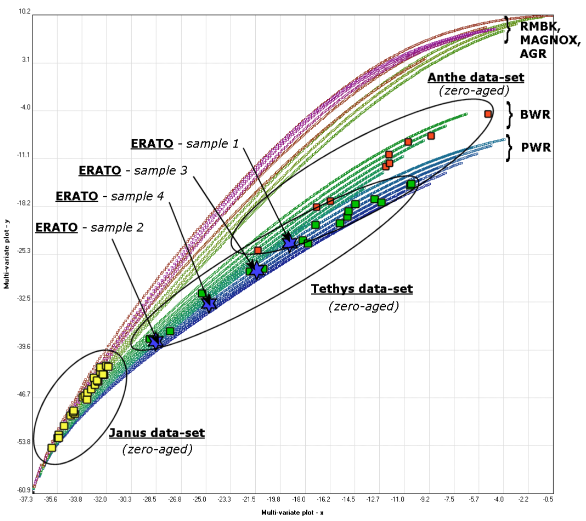


Figure 15. PuC diagram showing the Tethys, Anthe, Janus and Erato datapoints superimposed on the in-built library trajectories



LDA of the U-235/Total U vs. Total Pu/Total U data is able to comprehensively discriminate between Anthe and Tethys. As can be seen in Figure 13, LDA enables 100 percent correct classification of both Anthe and Tethys training data sets. LDA also identifies the four Erato datapoints as being consistent with the Tethys data set. The same conclusions can be made from LDA of the U-235/Total U vs. Pu-241/Pu-240 correlation (Figure 14), which also allows for 100 percent correct classification of the reactor data. Other isotopic correlations identify Erato as being consistent with the Tethys reactor. Percentage correct classification scores as calculated from the LDA Confusion Tables are provided in Table 3 and provide an indication of which isotopic correlations best provide the capability to distinguish between the two PWR reactors.

From the radiochemistry and isotopic correlations, it can be concluded that the Erato seized fuel pin is consistent with material from the Tethys reactor. The level of confidence was adjudged to be suggestive positive from the uncertainties on the regression lines and the similarities between the ranges of abscissa and ordinate values for the Tethys and Erato data sets in the isotopic correlation plots presented previously. A higher level of confidence was not ascribed as there is a need to better understand isotopic correlations involving Am-241 and the differing initial U-235 enrichment. Anthe and Janus were both adjudged to have a suggestive negative level of confidence for correlation with Erato. A further potential source of uncertainty is whether the seized fuel pin could correspond to operating conditions in another reactor for which the UK had not been given data in the exercise, especially due to the different initial U-235 enrichment. For instance, the Janus material data are notable for their low burnup. Higher burnup material from the Janus reactor would be closer to the Anthe or Tethys reactor data, and reduce the effectiveness of the isotopic correlations at discriminating between the different reactors.

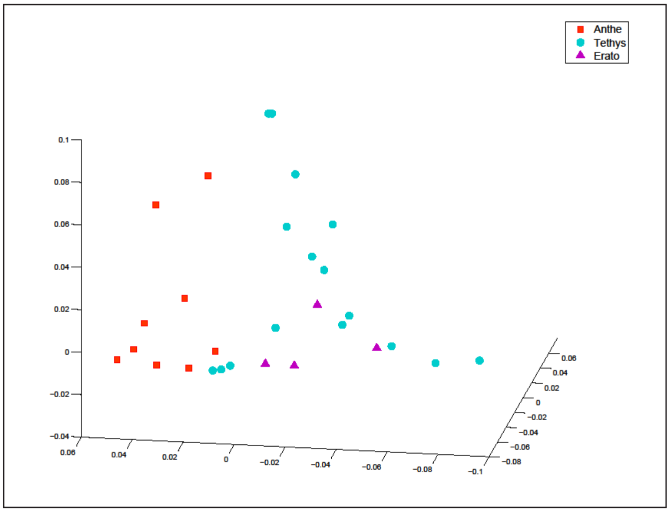
Reactor Type Multivariate Analysis (NNL)

NNL used a program called PuC for this desktop exercise. PuC was a pre-existing code that was developed as a tool to determine the origin of unknown plutonium samples that uses only the measured plutonium isotopic vector (Pu-238, Pu-239, Pu-240, P-241, Pu-242, and Am-241). PuC is based on a multivariate plot that provides maximum discrimination between different reactor types. The multivariate plot was originally arrived at using plant data from reprocessing operations, where the arbitrary linear coefficients applied to the plutonium isotopic vector were optimized to separate the different reactor types in the plotting space. A library of pre-calculated evolutionary tracks was generated for different reactor types and initial U-235 enrichments, using a multi-group lattice program. The evolutionary tracks show how the plutonium isotopic vector evolves as a function of burnup and provide a set of known reference against which a sample can be compared. PuC overlays the sample data on the plot and provides various graphical tools that the user can use to refine the assessment. This approach can be used to identify the reactor type and estimate the discharge burnup of the sample, as well as the initial enrichment. The built-in library covers all the major commercial reactor types worldwide.

Figure 15 shows the library evolutionary tracks with the Janus, Tethys, Anthe, and Erato datapoints superimposed on them. All the tracks originate from the origin at the bottom left, which corresponds to zero burnup, at which point the trace amounts of plutonium produced will only contain Pu-239. As the burnup increases, the minor isotopes build up and produce the divergent evolutionary tracks shown.

Using its in-built graphics functions, PuC was used to assign the best fit for each Erato sample to the in-built library. Table 4 summarizes the results. In each case, PuC indicates that the best fit is to a PWR, with the estimated discharge burnups ranging from 12 to 28 GWd/t and the initial enrichments ranging from 3.5 to 4.0 w/o. These estimated discharge burnups are very close to those from the Nd-148 measurements for Erato samples 1 and 2. However, for Erato samples 3 and 4 the PuC burnups are slightly higher than the Nd-148 measurements indicate.

Figure 16. LE visualization of the Anthe, Tethys, and Erato samples based on the features Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, U-234, U-235 and U-238



Examination of Figure 15 shows clearly that the Janus datapoints are distinct from the others and that the Erato points do not match them. Though the Tethys and Anthe datapoints overlap to some extent, the Erato points clearly fit best with the former. Interestingly, although the Anthe data are known to originate from a PWR, they fit more closely with the BWR evolutionary tracks. The explanation for this anomaly is not evident, but it is notable that some early PWR cores shared some features such as large water gaps and cruciform control rods that are now only found in BWRs. Table 5 identifies which Tethys datapoint most closely matches each Erato datapoint. In some cases, it was difficult to differentiate between two Tethys samples, hence why two matches are given in the table.

The final conclusion from PuC is that the Erato datapoints best fit Tethys, though there is some overlap with Anthe.

Table 3. LDA of selected isotopic correlations

			Erato fuel pin reactor prediction				
X – Value	Y-Value	% True reactor classification	1	2	3	4	Comments
Total Pu / U-238	U-235 / Total U	100%	Tethys	Tethys	Tethys	Tethys	
U-235 / Total U	Total Pu / Total U	100%	Tethys	Tethys	Tethys	Tethys	
Pu-238 / Total Pu	Pu-241 / Pu-240	100%	Tethys	Tethys	Tethys	Tethys	
U-235 / Total U	Pu-241 / Pu-240	100%	Tethys	Tethys	Tethys	Tethys	
Pu-240 / Total Pu	Pu-241 / Total Pu	100%	Tethys	Tethys	Tethys	Tethys	
Pu-240 / Total Pu	Pu-242 / Total Pu	96%	Tethys	Tethys	Tethys	Tethys	
Pu-238 / Total Pu	Pu-240 / Total Pu	96%	Tethys	Tethys	Tethys	Tethys	
Cs-137 / Total U	Pu-242 / Pu-241	92%	Tethys	Tethys	Tethys	Tethys	
Np-237 / Total U	U-235 / Total U	85%	Tethys	Tethys	Tethys	Tethys	
Pu-238 / Total U	U-235 / Total U	84%	Tethys	Tethys	Tethys	Tethys	
Pu-238 / Total Pu	Pu-242 / Pu-240	68%	Tethys	Tethys	Tethys	Tethys	
Cs-137 / Total U	Total Pu / U-238	64%	Tethys	Tethys	Tethys	Tethys	All Anthe samples are identified as being Tethys – poor discrimination.

Figure 17. LE visualization of the FISPIN and Erato samples based on the features Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, U-234, U-235 and U-238

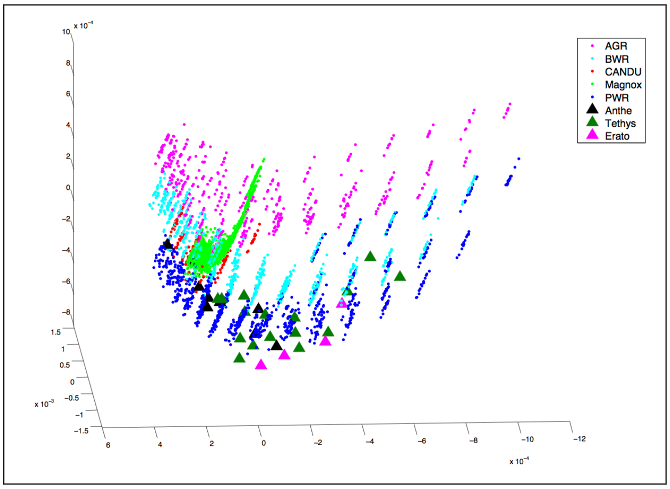


Table 4. PuC best fits to the Erato samples

ERATO sample	Most probable reactor type	Years between discharge and analysis	Estimated sample discharge burnup sample (GWd/t)	Estimated Initial ^{235}U enrichment (w/o)
ERATO-1	PWR	1.5	27.9	3.5
ERATO-2	PWR	3.5	12.2	3.5
ERATO-3	PWR	1.5	25.7	4.0
ERATO-4	PWR	1.0	19.0	4.0

Table 5. Closest matches for each Erato sample

ERATO sample	Closest match (1)	Closest match (2)
ERATO-1	Tethys-12	Tethys-5
ERATO-2	Tethys-1	Tethys-6
ERATO-3	Tethys-2	Tethys-7
ERATO-4	Tethys-11	n/a

Laplacian Eigenmaps (University of Liverpool)

The reactor data sets and seized Erato sample were extracted from their respective raw file formats using custom Matlab scripts and transferred into the Matlab environment (Mathworks Inc.) in matrix form. Principally our investigation has focused on visualization of the data set using an unsupervised spectral reduction method called Laplacian Eigenmaps (LE) to facilitate dimensional reduction. Unlike other dimensional reduction methods that are based on finding orthogonal directions that maximize data variance, LE will preserve characteristics of the original high-dimensional feature space, such as pairwise proximity information between the original samples.

LE works by converting the $n \times d$ data matrix X to an $n \times n$ similarity matrix $W=[w_{ij}]$, where w_{ij} represents the similarity between the i_{th} and the j_{th} rows of X, that is samples x_i and x_j. We have used Gaussian similarities between samples with a variance set to the median of the distances to measure the similarities for the matrix. The samples are then projected nonlinearly to a lower dimensional space of k dimensions where original d-dimensional samples x_i correspond to k-dimensional embeddings z_i. This is achieved by minimizing the sum of weighted pairwise distances between all n embeddings.[3,4] To facilitate visualization we have projected our eight original features representing the sample measurements of Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, U-234, U-235, and U-238 and projected these to a three-dimensional embedding. The three dimensional embedding has then been plotted to provide a visual representation of the data set.

The Janus reactor design has been eliminated from analysis based on the radiochemistry and isotopic correlations discussed previously. As there are no features consistent between all sample groups it has been necessary to exclude the Janus samples from further analysis. The Erato, Anthe, and Tethys samples have been visualized using LE dimensional reduction of the eight features that are common between these sample groups (Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, U-234, U-235, and U-238) and the results from this analysis can be seen in Figure 16. It is immediately apparent that three of the samples from the Erato pin are very closely grouped and the fourth sample appears to be somewhat of an outlier. The proximity of the cluster of Erato samples to the representative set of Tethys datapoints suggests that the sample would be a more likely candidate for the Tethys reactor. It should also be noted that although its nearest neighbor is a Tethys sample, the outlier Erato sample is near to a number of Anthe datapoints. Based on this analysis, it can be concluded that the seized Erato sample could have originated from the Tethys reactor. Similarly, this analysis supports a conclusion that the seized Erato sample could not have originated from the Anthe reactor.

In addition to the Anthe, Tethys, and Janus samples, our analysis has taken advantage of a representative data set of 3,682 samples generated using the FISPIN depletion codes.[5] Each sample represents the calculated isotopic composition of a given material with a variation of five different characteristics, reactor design, enrichment, irradiation, rating and cooling period. Figure 17 shows the visualization of the four Erato samples plotted with the FISPIN data set using LE dimensional reduction of eight features (Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, U-234, U-235, and U-238). Samples from the FISPIN data set have been categorized into the five representative reactor designs of AGR, BWR, Magnox, CANDU, and PWR, as shown in Figure 16. Based on this visualization we can conclude that the Erato samples would likely have originated from a PWR reactor design as the four Erato samples fall well within this region of the feature space. However, it should be noted that this investigation has the limitation of only being able to determine the reactor design within the confines of the class labels that we have available, namely the five reactor designs of this data set.

Further to our visualization work the data set has been classified using a classification method called Parzen window. The posterior probability is estimated from the data using the Parzen window technique, whereby a window function is employed for counting the number of samples that fall within a region of fixed volume and the class likelihood can be calculated.[3] The Parzen classifier has been implemented to determine the class of the Erato samples using the same data sets that were used for the visualization. The outcome of this classification has been used to quantitatively determine the most likely class of the Erato samples. As with the visualization the FISPIN data set has been used to determine the reactor design and the Anthe and Tethys data sets were used to determine the most likely reactor for this exercise. Classification with the FISPIN data set has positively identified the Erato samples as a PWR reactor. Based on the visualization work in Figure 17 this clearly supports the identification of the samples as a PWR reactor source. Furthermore, the classifier has identified the Erato samples as nearer to the Tethys reactor when compared with the Anthe samples, supporting the outcome from visual analysis of Figure 16.

From analysis of the classification work and visualization it can be concluded from our investigation that the Erato sample would have originated from a reactor with PWR reactor design and the most likely candidate reactor is that of the Tethys data set. Notably, this investigation has assumed no prior knowledge from the field of nuclear forensics and has been based solely on the application of pattern recognition techniques that could be widely used without the need for and in-depth knowledge of nuclear reactions and compositions. Our approach allows one to determine details about a given sample quickly, based on access to and expertise in the LE methodology. Such an approach might prove advantageous, especially where conclusions are needed promptly. However, it should be noted that there is a strong reliance on having a good set of representative training data such as those used in our investigation.

Conclusions at the End of the Exercise

The UK team found that the seized reactor pin, Erato was consistent with a PWR reactor pin. Based only on the data provided, the seized reactor pin was most consistent with the reactor Tethys. This allocation was made by three different approaches. The level of confidence was adjudged to be suggestive positive due to the difference in initial fuel enrichment and the need to better understand isotopic correlations involving Am-241. Comparisons between Erato and the other two reactors, Anthe and Janus, were at a suggestive negative confidence level. The possibility should be acknowledged that the seized fuel pin data may well be consistent with other PWR reactors, especially those built to similar designs around the world. The assumption is made that the reactor data correspond to the operating conditions employed at each reactor. Materials produced from unusual operating conditions may not correlate with the isotopics provided for each reactor, and the effectiveness of examining isotopic correlations will be diminished.

Comments Subsequent to the Revelation of the Scenario

Siarnaq was the source of the seizure in the Galaxy Serpent exercise. This reactor was not amongst the reactors supplied to the UK team. The UK team considered that the seized fuel pin was consistent with the Tethys reactor with a suggestive positive level of confidence. The main discrepancies identified between Tethys and the seized fuel pin were the differing initial fuel enrichments and the lack of correlation for plots involving Am-241. Whilst a suggestive positive level of confidence was expressed that the seized sample was consistent with Tethys; it was also stated that the data may well be consistent with other PWR reactors. Indeed, like Tethys, the Siarnaq reactor is another PWR; a comparison between the seized fuel pin, Siarnaq and Tethys would be a valuable future test of our methodologies.

The UK conclusion that Erato was not Janus or Anthe was correct.

The fact that Erato was not Tethys illustrates the problems that would be found in a real nuclear forensic case:

1. Each fuel rod in a reactor has a slightly different irradiation history, and there may be several different initial enrichments used in a core loading, and indeed over the reactor's operational life.
2. If a fuel rod comes from a different reactor than that which one has data for, due to the laws of physics, irradiation conditions and effects tend to produce similar ratios; thus,

although it may be possible to differentiate reactor types, it is very difficult to state categorically that a sample came from a specific reactor if the choice is between two similar reactors.

Thus, the UK conclusion was suggestive positive for Tethys: Tethys and Erato appeared to be similar, but not identical. Following the approaches made in this paper, the belief that Erato may have been Tethys would have been further investigated. Presumably at this later stage, it would be found that Tethys never had 3.5 percent fuel in it and would then have been eliminated. There would be no need to inspect Janus and Anthe records.

Such data set comparisons are thus only the first stage of a nuclear forensics investigation. For a country with a large number of reactors, the analysis may allow those of interest to be selected, especially if the country has many different reactor types, e.g., the UK. However, if the country has many reactors of similar designs, e.g., France, defining which reactor is the source of such a seizure would be difficult.

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The Hungarian Experience in the Galaxy Serpent Tabletop Exercise on National Nuclear Forensics Libraries

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Abstract

The Centre for Energy Research, Hungary, participated in Round 3 of the Galaxy Serpent virtual tabletop exercise (TTX). In the round of this TTX, four samples from a hypothetical spent nuclear fuel (SNF) seizure had to be identified using databases originating from three reactors based on SFCOMPO, an existing public domain international database of spent nuclear fuel originated from light-water reactors. The Hungarian Team created a model national nuclear forensic library (NNFL) using correlations of isotopes/total U vs. burnup. These correlations were treated as unique for each reactor and would be relevant for identification. Comparison for SNF was applied to determine whether or not the seized material was consistent with the data in the NNFL. Some conclusions are given regarding the origin of the seized samples using the method.

Introduction

In the context of the Galaxy Serpent virtual tabletop exercise (TTX) samples from a hypothetical spent nuclear fuel (SNF) seizure had to be identified using databases originating from three reactors based on SFCOMPO, an existing, public domain international database of spent nuclear fuel. Originally, SFCOMPO contains isotopic compositions for fourteen light-water nuclear reactors, seven pressurized water reactors, and seven boiling water reactors, from four countries (Germany, Italy, Japan, and the United States).[1]

Isotopic composition of spent nuclear fuel (SNF), obtained in post-irradiation experiments (PIE) is one of the important components required in the validation of burnup credit methodologies. It is the basis for the evaluation of several properties of SNF, including decay heat, radiation dose, and the neutron multiplication factor of any configuration containing spent fuel. Since the determination of this isotopic composition is usually carried out using calculation tools, the validation of these tools is an important aspect to be taken into consideration.[2]

SFCOMPO data is most useful in developing isotope correlations corresponding to the reactors. These isotope correlations are generally expressed as isotopic ratios versus a surrogate for exposure time in the reactors (e.g., burnup, build-up of Pu, etc.). Using such correlations, it is anticipated that mathematically well-behaved patterns can be constructed for each reactor, and these correlations will identify differences between reactors or classes of reactors. In the frame of the Galaxy Serpent TTX, two tasks had to be carried out: creating a model national nuclear forensics library (NNFL) from provided SFCOMPO information, and using the NNFL to determine whether data provided from a hypothetical seized material are consistent or not with any of the materials described in the NNFL developed in the first phase.[1]

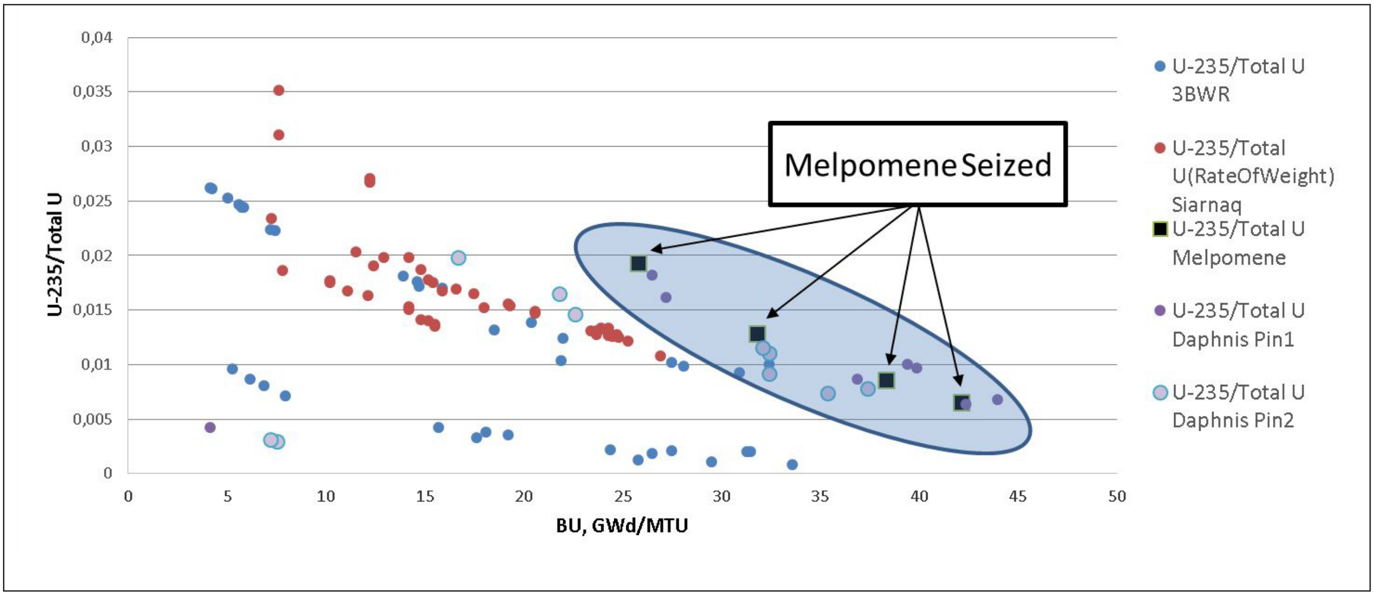
The NNFL created by the Hungarian team was based on the relationship between the ratios of isotopes/total U and burnup. The correlations of isotopes/total U vs. burnup were treated as unique for each reactor and therefore would be relevant for identification. Comparison for SNF was applied to determine whether or not the seized material was consistent with the data in the NNFL.

Method and Data

Creating a National Nuclear Forensic Library

As described in the introduction, the isotope correlations can be obtained as isotopic ratios versus a surrogate for exposure time in the reactors (e.g., burnup, build-up of Pu, etc.). Using such correlations it is possible to identify differences between reactors or classes of reactors. In this work the burnup was used for creation of classes from reactors and to find the origin of hypothetically seized SNF samples. The basic assumption is that neutron fluence distribution in the reactor zone is different in every reactor. Therefore, using burnup and various isotope correlations as well as initial enrichment as parameters of the nuclear fuel characteristic data and distinct classes can be obtained from the data of reactors. As described in References 3 and 4, the content of Cs-137 in a module vs. burnup, the ratios

Figure 1. The ratio of U-235/total U vs. burnup of the Melpomene seized samples in the Hungarian NNFL



of U235/total U and total Pu/total U vs. burnup were treated as unique for irradiated material originating from 3.6 percent U235 enriched fuel assemblies in reactor VVER-440 at Paks Nuclear Power Plant. These correlations were considered for calculating the content of U-235 and U total and Pu total in mixed irradiated materials.

In this work, in the frame of the Galaxy Serpent TTX, the content of an isotope vs. burnup in unit mass of irradiated material were treated as unique for each type of reactor. Therefore these relationships were considered as an available basis for creating NNFL, then for identification of seized samples. The NNFL created by the Hungarian Team from three different reactors (Daphnis, Enceladus [3BWR], Siarnaq) has three sets of following ratios of isotopic/total U vs. burnup: ^{234}U/Total U, ^{235}U/Total U, ^{236}U/Total U, ^{238}Pu/Total U, ^{239}Pu/Total U, ^{240}Pu/Total U, ^{241}Pu/Total U, ^{242}Pu/Total U, ^{241}Am/Total U, ^{242}Am/Total U, ^{137}Cs/Total U, ^{237}Np/Total U vs. burnup. Mathematically, here the total U in ratio of the isotopic/total U was a normalized factor for unit mass. In the first approaches, the value of the total U was not changed during irradiated time.

In the TTX, three databases from three different reactors (Daphnis, Enceladus (3BWR), Siarnaq) were provided in spreadsheets (Excel files). While the NNFL created by the Hungarian team were temporarily represented in twelve figures, in each figure there were ratios of one type isotopic/total U vs. burnup of three reactors (Daphnis, Enceladus (3BWR), Siarnaq).

Identification of Hypothetical Seized Material

The names of seized samples were Melpomene 1-4. For identification of each seized sample, the value of isotopic/total U vs. burnup was inserted into corresponding figure of NNFL. Comparison was applied to determine whether or not the seized material was consistent with the data in the NNFL. If the data of a seized sample corresponds to the NNFL data of two or more reactors, then the comparison must be continued using the data of another isotope. The use of more isotope characteristics for comparison would increase the confidence level of identification.

Results and Discussion

The results of our investigation can be seen in Figures 1-4, where the data of the seized samples were inserted into the Hungarian NNFL.

Figures 1 and 2 show Melpomene seized samples on NNFL of ratios of U-235/total U and Pu-241/total U vs. burnup, respectively. They were two among twelve figures of NNFL created by the Hungarian team for three reactors (Daphnis, Enceladus [3BWR], Siarnaq), in which the data points and distinct classes can be seen among the reactors.

Figure 3 shows the ratio of U-234/Total U vs. burnup of the Melpomene seized samples in the NNFL (but without data of Siarnaq reactor). It can be seen that data points result in most of the cases distinct groups or classes among the reactors. Moreover in the case of the Daphnis reactor, two individual groups

Figure 2. The ratio of Pu-241/total U vs. burnup of the Melpomene seized samples in the Hungarian NNFL

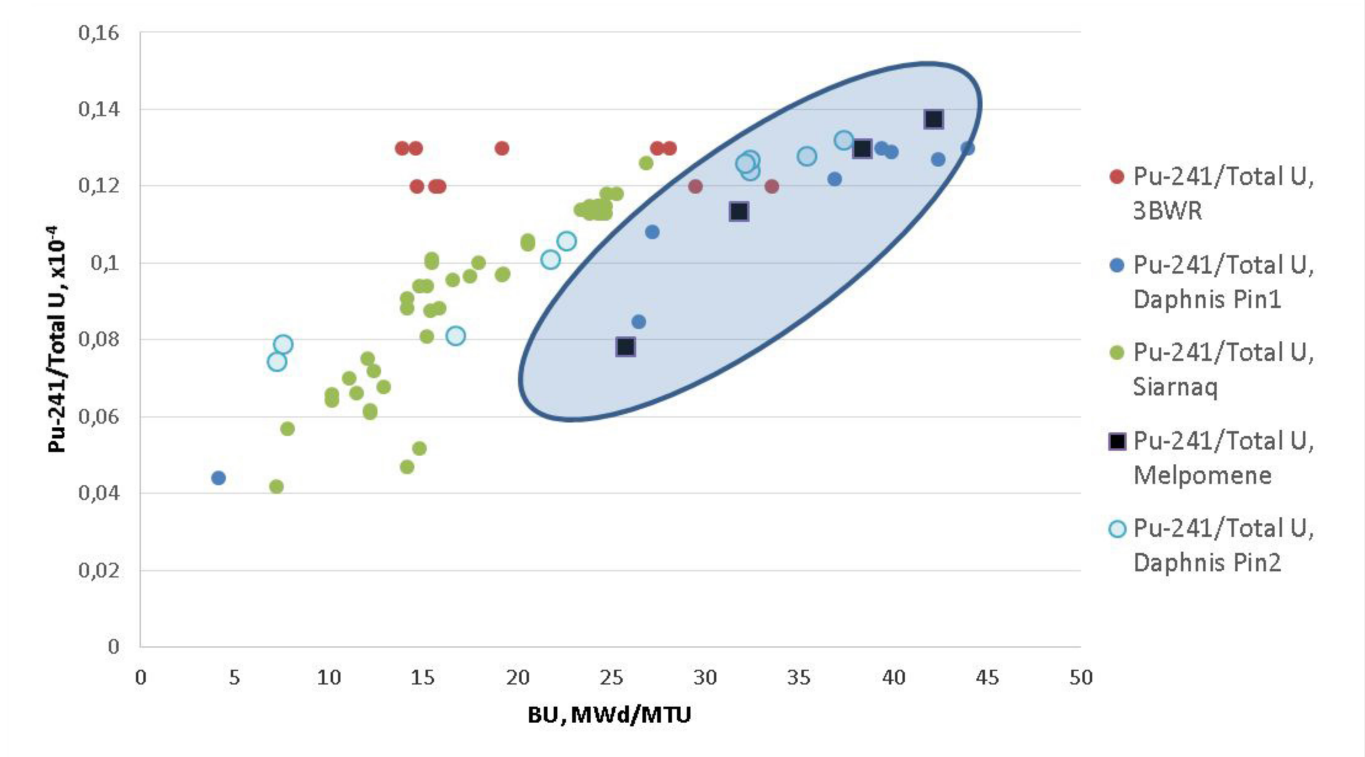


Figure 3. The ratio of U-234/total U vs. burnup of the Melpomene seized samples in the Hungarian NNFL

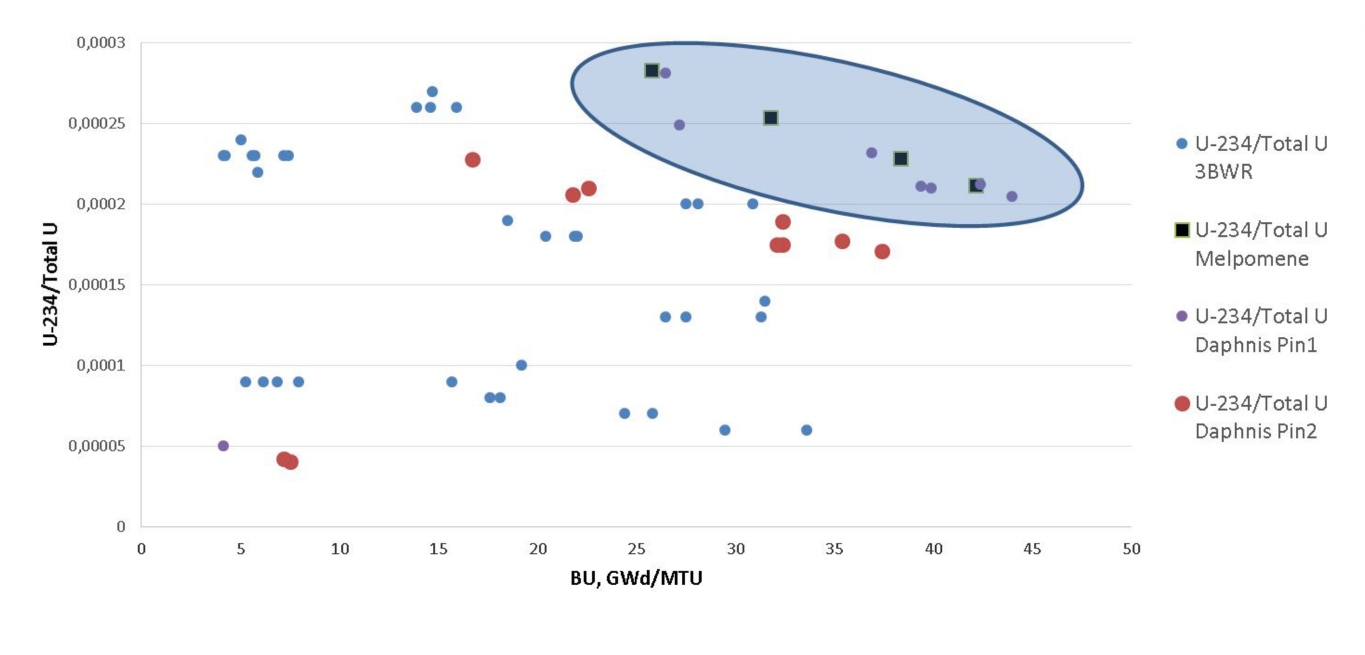
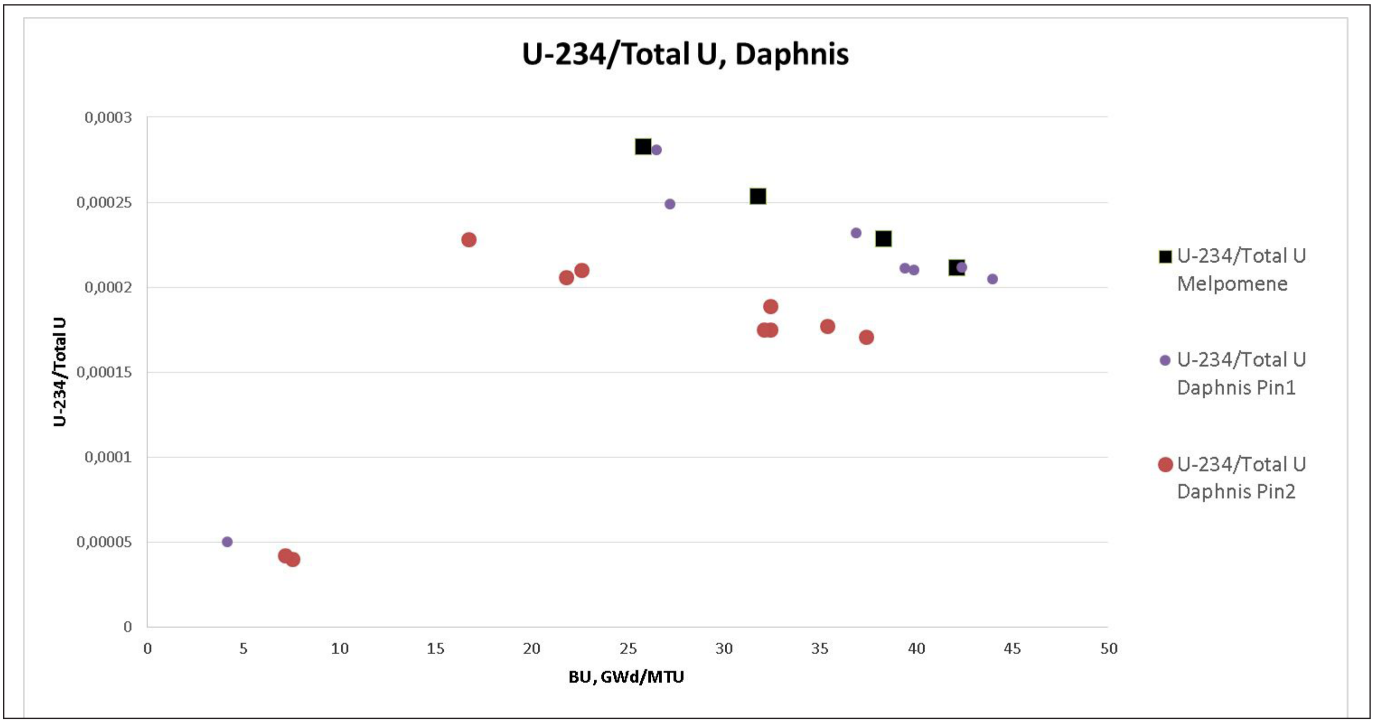


Figure 4. The ratio of U-234/total U vs. burnup of the Melpomene seized samples in the Hungarian NNFL (Zoom in Reactor Daphnis Pin 1)



can be found within the reactor. Considering the fitting of the data points of the seized samples it is seen that they are consistent with the data points of one group of Daphnis reactor (Pin 1).

Figure 4 illustrates the individual groups of Daphnis reactor (Pin 1 and Pin 2) and the fitting of the data points of the seized samples into Daphnis Pin 1 group.

Conclusion

In the framework of the Galaxy Serpent virtual tabletop exercise, four spent nuclear fuel samples from a hypothetical seizure had to be identified using databases and different other parameters originating from three reactors based on SFCOMPO open-source international database. The Hungarian team used the burnup as main characteristic parameter of SNF to create a NNFL for determining the origin of the hypothetically seized SNF samples. Investigating different isotope correlations as functions of burnup, different classes of reactors were separated. Based on the analysis of the relevant parameters of the seized samples all of the four items could be associated with the material from reactor Daphnis. For reliable conclusions further investigations would be necessary to analyze more characteristic parameters and use mathematical and statistical evaluations. It is also important to note that for such kind of investigations the knowledge of reactor operation and spent fuel characteristics is also important together with well-used statistical evaluations. Organization of such kind of virtual tabletop exercises in other topics (e.g., with fresh fuel data) would be also highly useful for the establishment of NNFL's in individual countries.

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U and Pu Isotopic Correlations to Check Consistency of Seized Nuclear Material Against a Known Inventory

Team Sweden Effort to Create and Use a Virtual National Nuclear Forensic Library (NNFL) in the Galaxy Serpent Tabletop Exercise

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Abstract

As a part of the Galaxy Serpent Tabletop Exercise, a rudimentary notional national nuclear forensic library (NNFL) was created. The NNFL was based on irradiated fuel sample analysis data provided by the exercise management. It was then used to evaluate data from a notional seized material. Our effort was concentrated on a small number of uranium and plutonium isotopic ratio correlations to draw conclusions on the consistency of the seized material data against data in the NNFL to answer the question if the seized material could have originated from the inventory described by the NNFL. The isotopic relationships used were selected based on several criteria, including known high discriminatory power in most cases involving irradiated uranium fuel, universality (data available for all materials in the NNFL and for the seized material), constancy (no short half-lives involved) and low measurement uncertainty. Our analysis resulted in a clear negative conclusion: the unknown sample data did not match any material in the notional NNFL. In addition, problems and possible strategies foreseen for setting up a real NNFL in a state with a comprehensive nuclear fuel cycle over many decades are discussed, stressing the central role of operating and accountancy records in conjunction with fuel depletion modeling to make maximum use of costly sampling and analysis in mapping a complex nuclear material inventory into a useful NNFL.

Introduction

Nuclear decay and technological processes such as enrichment and neutron irradiation cause regular and predictable changes in the isotopic composition of nuclear material. Therefore, precise and accurate isotopic data can be used to form conclusions about the history and identity of such material. Various techniques based on measurement and analysis of isotopic relationships can be used in nuclear safeguards where examples range from verification of operator declarations concerning nuclear material[1,2,3] to environmental sampling techniques aimed at detecting undeclared nuclear material and activities.[4,5,6] Another application, the focus of this work, is forensic investigation aimed at establishing possible connections of a seized specimen outside of regulatory control to known inventories of nuclear material.

Although access to precise and accurate measurement data for judiciously chosen parameters obviously form a basic prerequisite for any nuclear forensic capability, correct and efficient evaluation of data is also critical. The information, tools and methods and above all the skill sets required need to be identified and maintained. Therefore, reaching out to the adequate subject matter experts (SME) is important. Such SME could be persons with extensive knowledge of, e.g., different parts of the nuclear fuel cycle.

The range of analytical techniques available for post-irradiation analysis of nuclear material, and the range of measureable parameters produce a potentially large dimensionality of the resulting data set. Multivariate statistical approaches have been explored in order to compress the dimensionality and make optimum use of the available information.[7,8] Alternatively, a small number of specific relationships known from experience and from nuclear physics and engineering principles to provide powerful discrimination regarding properties of interest, such as fuel enrichment, power history or reactor and fuel type, may be selected. The latter approach has been used in this work.

Theory

Irradiation of uranium fuel in a nuclear reactor causes depletion of the fissile isotope ^{235}U and buildup of heavier uranium isotopes and other actinides: ^{236}U and progressively heavier isotopes of neptunium, plutonium, americium, and curium. Obviously, many other nuclides are available for analysis of this and other nuclear and radiochemical processes. However, the discussion in this short paper will mainly be limited to analysis based on actinide isotopic ratios and their correlations in irradiated uranium fuel. Besides being most relevant to the task presented by the Galaxy Serpent Tabletop Exercise (TTX), actinide isotopic data generally have the advantage of being more directly specific to the identity and provenance of a material. Pragmatically, data available from sample analysis are often, due to the measurement methods used, more precise and accurate for the major actinides (uranium and plutonium) than for e.g., the fission products, particularly if the sample is of unknown origin.

The build-up rates of actinides depend on the interplay of neutron reactions causing capture or fission, and nuclear decay of short-lived nuclides at intermediate steps. The capture and fission reaction cross sections for each nuclide in a build-up chain vary with the neutron spectrum, which in turn is determined by details in reactor and fuel design such as the isotopic composition of the fuel and the type of moderator and its distribution relative to the fuel. The build-up of nuclides in chains that contain short-lived members is also influenced by the relative rate of neutron capture reactions and nuclear decay of those members, which therefore introduces a dependence on the neutron flux, a reactor operating parameter.

Use of Uranium and Plutonium Isotopic Correlations to Identify Nuclear Material

The ratios of uranium and plutonium isotopes in spent uranium fuel are easily measured and useful signatures for evaluating similarities and differences between different fuels. Specifically, ^{235}U is depleted though fission and neutron capture to produce ^{236}U, and ^{239}Pu and heavier isotopes of plutonium are created through neutron capture on ^{238}U. The ratios between uranium and plutonium concentrations and isotopic compositions change with irradiation exposure (or burnup), and follow predictable trajectories. These trajectories change shape based on initial fuel composition, the neutron spectrum the fuel is exposed to, and the irradiation history of the fuel. The build-up of ^{238}Pu (by neutron capture in ^{236}U, followed by decay to ^{237}Np, a second neutron capture and finally decay to ^{238}Pu) is also sensitive to the neutron flux, since it is governed by the relative rates of decay and neutron capture in the short-lived isotope ^{237}U.

After irradiation, the process of nuclear decay will cause additional changes to the isotopic composition of the material. Among the major actinides, a process often of interest in nuclear forensics is the beta decay of ^{241}Pu to ^{241}Am. The half-life of 14.3 y means that in some circumstances a plutonium containing material can be dated on the basis of the ^{241}Am/^{241}Pu ratio. [9] Lack of knowledge of the initial (end-of-irradiation) ^{241}Am concentration places a limit on the achievable accuracy, and in relative terms this limitation will be more severe for recently irradiated material. The initial ^{241}Am concentration can be estimated with varying accuracy by modeling the irradiation process, provided other information (e.g., a burnup estimate) is available to constrain the modeling.

Modeling is often useful in order to illuminate and understand the systematic effects in isotopic correlations in known materials, and to test hypotheses concerning the provenance of a material of unknown origin. A number of depletion codes of varying sophistication exist to model the effects of neutron irradiation of e.g., uranium fuel.

Method

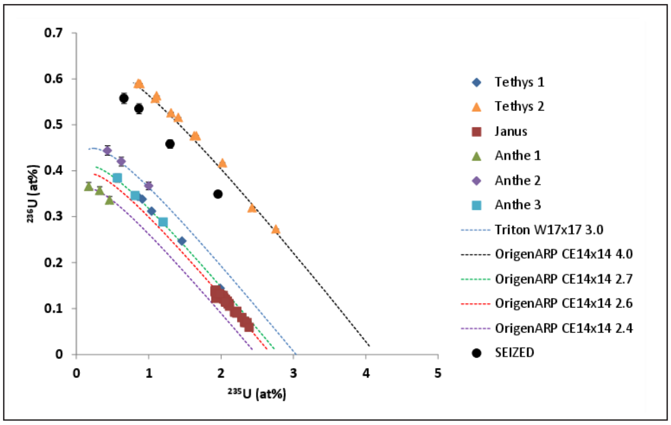
For the Galaxy Serpent TTX, we received and incorporated into a rudimentary national nuclear forensic library (NNFL) a data set comprising measurement results for a total of fifty-six samples from three different reactors: nine, sixteen, and thirty-one samples from Anthe, Tethys, and Janus reactors, respectively. A total of eighty-one different parameters were represented in the data set, counting each ratio as one parameter only, even though many were provided both as atom ratios and as weight ratios. Only nineteen parameters were provided for all samples. In order to first compare all sample data on an equal footing where possible, initial efforts were focused on this subset of nineteen parameters, which included uranium and plutonium isotopic abundances and ratios, elemental ratio of uranium and plutonium, and fission-product based burnup estimates. In addition, ^{241}Am concentrations, while not provided for all samples, were nevertheless included in the initial structure of the NNFL when provided. Among the parameters not used were actinide concentrations (including some minor actinides) as well as various fission and activation product concentrations and activity ratios.

The rudimentary NNFL consisted of a set of Excel spreadsheets containing uranium and plutonium isotopic abundances and isotopic ratios as well as ^{241}Am concentrations for each group of samples, including data from any unknown, seized material. An initial subdivision of the Anthe, Janus, and Tethys groups of sample data into three, one, and two different materials, respectively, was made based on differences in initial uranium enrichment evident from plotting the ^{236}U isotopic abundance versus the ^{235}U isotopic abundance (see Figure 1). Each Anthe material (Anthe 1 – 3) was represented by three samples, Tethys material 1 by five and Tethys material 2 by eleven samples. Presumably, for a real NNFL, the owner of the material would be aware of the properties and origins of different materials in his custody and could make this useful grouping based on his own records.

An adjustable time-before-measurement was also introduced that allowed for decay correction of ^{241}Pu and ^{241}Am as well as for plutonium isotopic abundances due to the decay of short-lived ^{241}Pu. This was for the purpose of age-dating and for comparison to modeling results referring to a past point in time (e.g., the end of irradiation).

The SCALE fuel depletion code[10] was used to model irradiation of uranium fuel in a reactor. A set of irradiation scenarios was chosen that would roughly reproduce the uranium isotopic relationships in the NNFL. No particular effort was spent on scenario selection beyond specifying initial uranium enrichment to reproduce the data. The models chosen were simply the closest to hand: a combustion engineering (CE) 14x14 PWR fuel assembly model provided with the OrigenARP component of the SCALE package, and in one case a Westinghouse 17x17 PWR fuel design modeled in-house using the Triton module in SCALE. The scope of this modeling was very limited in this case, simply aiming to confirm and solidify basic understanding of the materials in the NNFL as originating from the irradiation of different low-enriched uranium (LEU) fuels in thermal reactors. It was also done in preparation for the potentially more challenging task of evaluating data from measurements on an unknown seized material. Generally, samples from such a seizure might be expected to show different levels of burnup than samples in the NNFL, but might nevertheless represent the same fuel material.

Figure 1. Uranium isotopic abundances for known materials in the virtual NNFL and for the seized material (black). Also shown are model trajectories from SCALE.

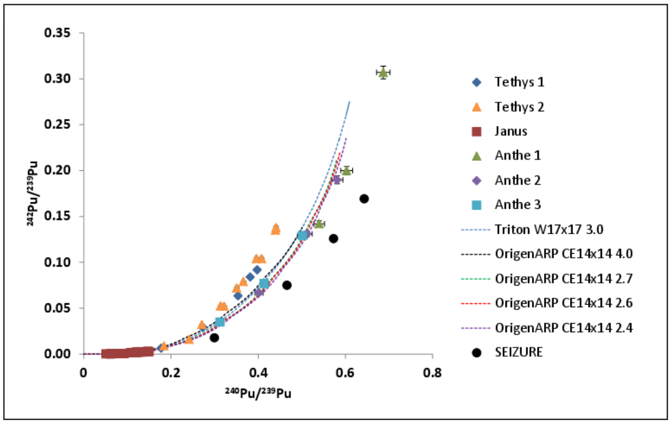


The seized-material data set received comprised data from four samples, each characterized by fifteen parameters including isotopic concentrations of uranium isotopes, plutonium isotopes, ^{237}Np, ^{241}Am, ^{242m}Am, ^{137}Cs, and ^{99}Tc as well as burnup estimate based on ^{148}Nd. Applicable parts of this data set were transformed to fit the structure of the rudimentary NNFL (i.e., isotopic abundances were computed from concentration data etc.) and compared to data for the known materials.

Results

Examining first some signatures of the six known materials in the NNFL (three Anthe materials, one Janus, and two Tethys materials), the uranium isotopic abundances shown in Figure 1 follow SCALE-computed burnup trajectories that would be expected for LEU fuels with various initial enrichment in the approximate range of 2.4 – 4.1 percent. The initial enrichment estimates are approximate since without additional information, we cannot know for certain what the ^{236}U content of the fresh fuel was. However, this is of no consequence for the task of discriminating between different materials. Clearly, samples have been collected from fuel with different burnup (i.e., results fall at different positions along each trajectory), probably corresponding to different positions within the core but possibly collected at different points in time in the operating history of fuel with a given enrichment. Lacking reference dates for the measurements, we avoid depending on the ^{241}Pu isotopic data by expressing Pu isotopic relationships as ratios between isotopes 239, 240 and 242 only. Figure 2 shows ^{242}Pu/^{239}Pu vs. ^{240}Pu/^{239}Pu isotopic ratios in the various samples and the trajectories expected for the same SCALE fuel depletion scenarios

Figure 2. Plutonium isotopic ratios for known materials in the virtual NNFL and for the seized material (black). Also shown are model trajectories from SCALE (same models as shown in Figure 1 for uranium isotopic abundances).



as shown in Figure 1. We note that the known materials group differently in the plutonium correlation plot: all three Anthe materials, with different enrichments, appear reasonably well-described by the fuel design model chosen more or less ad hoc for the SCALE calculation. The two Tethys materials, also with quite different enrichment, nevertheless follow a more or less common trajectory that is different from the selected SCALE model and from the Anthe materials. The uranium and plutonium isotopic data clearly give complementary information on the characteristics and origin of the various materials, as expected.

Turning to the task of discriminating the seized material from known materials in our inventory, we first compare the most basic isotopic correlation available for irradiated uranium fuel — the buildup of ^{236}U with the depletion of ^{235}U (Figure 1). It is evident that the seized material is on a different trajectory than any of the materials in the rudimentary NNFL (Figure 1), signifying primarily a different initial uranium isotopic composition, most likely (absent an unusually high initial ^{236}U level in the fresh fuel) a different initial enrichment.

As additional confirmation, the relative buildup of the heavier plutonium isotopes with irradiation exposure was used (Figure 2). This correlation depends more on the neutron spectrum than on initial fuel enrichment and therefore forms a useful complement to the uranium burnup trajectory, as discussed. The trajectory of the seized material is clearly separate from any of the NNFL materials.

Discussion

Although the data provided for the "seized material" in the Galaxy Serpent TTX could be discriminated from any of the "known" materials in our inventory by straightforward comparison of uranium isotopic abundances, it is interesting to discuss some possible avenues of analysis if both the uranium and long-lived plutonium isotopic correlations (Figures 1 and 2) had been less conclusive.

An obvious possibility is age-dating on the basis of the decay of ^{241}Pu. The required ^{241}Am data was available for the seized material, but only for the Anthe materials in the NNFL. Generally, to gain much information through the comparison of the ^{241}Am/^{241}Pu ratio from two measurements, reference dates for the two measurements are required. These were not provided for the TTX. However, on the reasonable assumption that the seized material was measured later than the known materials, one could have concluded from the much lower ^{241}Am/^{241}Pu ratios in the seized compared to the Anthe materials that the seized material had been irradiated much more recently than any of the known materials from Anthe. In fact, the ^{241}Am/^{241}Pu ratios in the seized material are so low that it is difficult to construct an irradiation and decay scenario that would allow more than about a year between end of irradiation and measurement. If the owner knows that all materials in an inventory are considerably "older" than that, the ^{241}Am data for the seized material would be a useful indicator that the seized material does not come from that inventory.

Other possible "tie-breakers" are available in the data for the seized material. They will be mentioned but not discussed in any detail since they were in fact not utilized and because the authors lack experience in using most of them for the purpose of identifying nuclear material. The concentrations of the fission product ^{137}Cs (half-life 30.1 y) and the actinide ^{242m}Am (half-life 141 y) are possible "clocks" in the data for the seized material, although the latter was not available for all materials in the NNFL. Again, some information on relative reference dates for measurements would be required to use any timing information obtained to best advantage. Concentrations of long-lived ^{237}Np and its relatively short-lived (88 y) activation product ^{238}Pu are sensitive to neutron flux since they are produced by decay of short-lived ^{237}U, in competition with neutron absorption. ^{238}Pu would be the more attractive of the two as the data are more precise and since it would allow use of in-element abundance ratios. However, in view of its short half-life, independent knowledge of its age and/or relative measurement dates would again be preferable. The fission product ^{99}Tc is long-lived (211500 y), but it is not clear that it should be expected to be sufficiently different between materials so similar that they cannot be distinguished on the basis of uranium and plutonium isotopic composition in the first place. Burnup estimates for the seized material were provided and could be useful if outside the range of burnup for material in the inventory, known to the owner from operating records and possibly supported by sampling.

The last point leads to the interesting problem of how an actual NNFL might be usefully and efficiently structured and populated in the case of a state with a large nuclear fuel cycle, having operated over many decades and resulted in a wide variety of material differing in all the variables discussed so far—age, fuel, and reactor design, operating history, initial isotopic composition and so forth. Clearly, a strategy would be needed that minimizes the extent of costly sampling, analysis and sample storage and optimizes the usefulness of such activities when performed. Tentatively, it would seem that an extensive survey of the most detailed operating and accountancy records available would be beneficial as a first step. The results would be a natural part of an NNFL in themselves, but would also guide limited "pilot" sampling and analysis of material identified by the survey as located at the extremes in the variables of interest. What the detailed set of variables of interest should be needs much further study, but it would certainly include enrichment and burnup, possibly for each reactor, for each major fuel design, for each fuel shipment received and so on. A next step might then be to model actinide, and probably fission and activation product isotopic concentrations for each fuel history. When models have been developed that reproduce actual sampling results sufficiently well, they would constitute a major tool in assessing whether an unidentified nuclear material came from the inventory described or mapped by the NNFL. Only quite limited additional sampling and analysis might be required to fill in critical gaps between extremes and further validate the models.

Conclusion

The Galaxy Serpent TTX task of distinguishing the "seized material" from some materials in a virtual inventory on the basis of the sample analysis data provided could be solved in a straightforward way by application of uranium and plutonium isotopic correlations that were known to have particular discriminating power in most cases involving irradiated nuclear material. If the uranium and plutonium isotopic correlations used had proved to be ineffective for accomplishing the discrimination, other data provided might have been of use, particularly burnup estimates and age-dating on the basis of ^{241}Am/^{241}Pu ratios. For a large nuclear material inventory, based on an extensive nuclear fuel cycle, mapping the entire realm of possible signatures solely by sampling and analysis could become a prohibitively cumbersome and expensive endeavor, unless guided by a well-conceived strategy. Such a strategy would most likely make extensive use of existing operating and accountancy records as well as of fuel depletion modeling. Ongoing and future work with development of the NNFL concept in different States will need to address some of these issues.

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The Galaxy Serpent Exercise: Methodology, Experience, and Findings of the Institute for Transuranium Elements

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Abstract

In the framework of the Galaxy Serpent exercise, a data set of an unknown spent fuel material (code name Terpsichore) was compared against three data sets associated with three distinct reactors. The data of the known materials were compiled in a model national nuclear forensics library. The comparative evaluation aimed at (i) assessing whether the hypothetical seizure is or is not consistent with material contained in the library, and, (ii) identifying the reactor the unknown material may be associated with. Two different evaluation methods were applied. The first one is based on the isotope correlation technique and concluded that one of the three known reactors (code name Siarnaq) as potential origin of the unknown material. The second method uses multivariate analysis for clustering and classification of the data, identifying the same known reactor (Siarnaq) as the likely origin of the Terpsichore material. Thus, the evaluation resulted in a consistent answer provided by two independent methods. In addition to demonstrating the usefulness of having a national nuclear forensics library, the exercise showed that selected statistical techniques, such as principal component analysis or factor analysis, may serve to optimize the content of a national nuclear forensics library by eliminating redundant (correlating) variables.

Introduction

In the framework of the Nuclear Forensics International Technical Working Group (ITWG) a cooperative tabletop exercise was conducted.[1] Participating laboratories were provided with data sets of spent fuel of known origin. Though the data had been taken from real samples (available through a publicly accessible database) the scenario was hypothetical and the reactors were given code names. The Institute for Transuranium Elements (ITU) was provided with data sets on spent fuel originating from three known reactors. Two of the reactors, Anthe and Siarnaq, were pressurized water reactors while the third reactor, Hyperion, was a boiling water reactor. In the hypothetical scenario of the present exercise a data set of an unknown material (originating from Terpsichore reactor) had to be compared against the known materials in order to determine whether the Terpsichore material is or is not consistent with one of the three known reactor materials. Participants were free in the choice of methodology for analysing and comparing the data.

In a first step the data of the known samples were compiled in a common format using a simple Excel spreadsheet. This repository of data served for the purpose of the exercise as the national nuclear rorensic library (NNFL). It should be noted, though, that the data provided for the known samples were not homogeneous, i.e., the samples did not share the same complete set of parameters. This applied also to the unknown sample. For example, the concentration certain fission products (such as ^{99}Tc) or actinides (such as ^{237}Np) were not available for every sample.

ITU decided to use a two-track approach. On the one side we used an isotope correlation technique, which made direct use of the (sometimes fairly small) difference in nuclear physics behavior of distinct reactor types. The differences are related to factors such as varying initial enrichment of the fuel or different neutron energy distribution. Correlating measurable parameters of the spent fuel may help to distinguish between different reactors. The second method we used was a statistical approach using a supervised technique. Both methodologies and the respective findings are described here in some detail.

Isotopic Correlation Technique

The first technique used for comparing the data of the unknown sample (Terpsichore) with the data in the NNFL, was the so-called isotope correlation technique (ICT). ICT is a method that has been examined earlier for its applicability in nuclear material safeguards, where it was used to check the consistency of the safeguards declarations of spent fuels as well as to replace some analysis by correlations.[2,3] The correlations were

Figure 1. Isotope correlation using the relative isotope abundances ^{235}U vs. ^{236}U

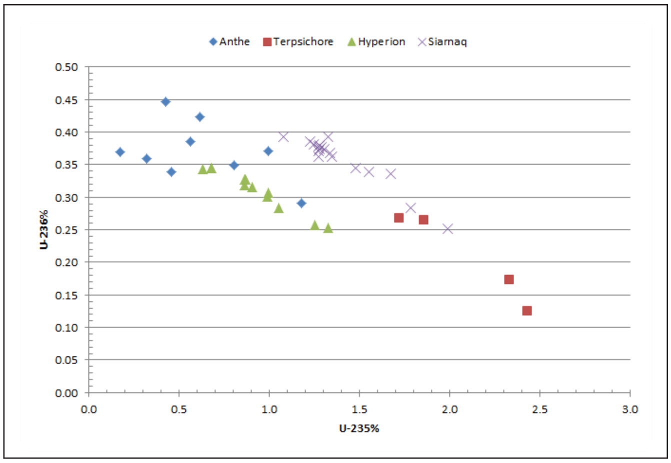
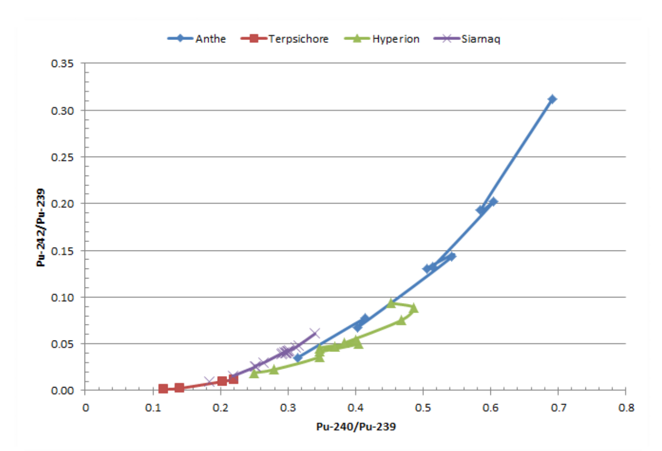


Figure 2. Isotope correlation using isotope abundance ratios ^{240}Pu/^{239}Pu vs. ^{242}Pu/^{239}Pu



typically two-dimensional and could thus be easily visualized using x,y diagrams. However, not all of the correlations that had been identified as useful through the Isotope Correlation Experiment[2] could be applied for Galaxy Serpent due to the incompleteness of data. The latter, however, has to be recognized as a realistic constraint in the use of NNFLs.

In the first step, we established the correlations that provide the best possible distinction between the three reactors contained in our NNFL. Typically such correlations reflect differences in the initial ^{235}U enrichment, neutron fluence, and neutron spectrum hardness (among others). In our case, the investigated correlations included uranium and plutonium isotopes. If the data set for the unknown sample had been more complete, other transuranium elements (Np, Am, and Cm) could also have come into consideration together with certain fission products.

In the second step, the data points of the unknown sample (Terpsichore) were inserted in the graphs. Some of the correlations we examined are illustrated in Figures 1-3. These were chosen, because they visualize the distinct separation of the three reactors from each other (Figures 1 and 2). Most of the correlations indicated inconsistencies for the Anthe data set, which showed a non-linear and sometimes a discontinuous behaviour as can be seen in Figure 1. Also data sets from other reactors (Hyperion and Siarnaq) showed occasionally non-linear behavior, though for fewer correlations (e.g. Hyperion data in Figure 2).

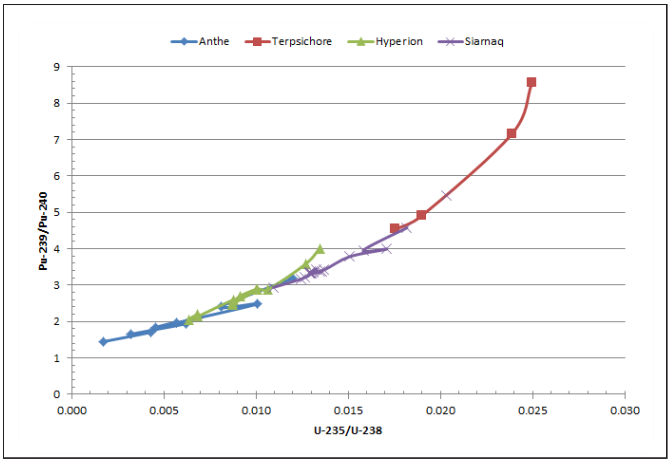
For visualization of the data and correlations some simplifications were used:

1. The uncertainties were excluded from the figures, because they were generally low compared to the scatter of data points (with a given reactor) and to the differences between the reactors.
2. ^{241}Pu was generally excluded from the ICT, because of the relatively short half-life of ^{241}Pu and the fact that the cooling time of the spent fuel was not known for most of the data sets.
3. In Figures 2 and 3 the data points have been arbitrarily connected by a solid line for better visualization. Due to the sometimes poor linearity or discontinuous appearance of data, an extrapolation outside the range of available data was not recommended.

Generally we observe in Figures 1-3 that the Terpsichore data points are largely outside the range covered by the other reactors. More specifically, the ^{235}U/^{238}U ratio in two out of four data points of Terpsichore is higher than in the three known samples (Figures 1 and 3). This might be due to higher initial enrichment or to a lower burnup of the unknown material. Also the ^{240}Pu/^{239}Pu and the ^{242}Pu/^{239}Pu ratios in two out of four data points in the Terpsichore data set are lower than in the other materials (Figure 2) indicating a lower burnup of the fuel.

The isotope correlations suggest the unknown material (Terpsichore) to be consistent with one of the known reactors (Siarnaq). Figure 3 shows a partial overlap of the curves established for these two reactors, while for Hyperion and Anthe the curves would need to be extrapolated to a degree which is neither recommended nor justified. The same applies for Figure 2. The correlation shown in Figure 1 allows a clear distinction between Terpsichore and Siarnaq on the one side and Anthe and Hyperion on the other.

Figure 3. Isotope correlation using ^{239}Pu/^{240}Pu vs. ^{235}U/^{238}U



Multivariate Analysis

If a NNFL is available, a more advanced approach, multi-variate statistics, can also be applied to help identify the origin of an unknown sample. Using this method, we can not only consider multiple variables at the same time, but we can also understand the relationship between the variables and interpret the measurement results with higher confidence. The multivariate analysis has the advantage that, due to the higher number of variables, it can generally cope with higher measurement uncertainties (both in the library data set and for the measured data of the unknown sample), redundant data or incomplete data sets in the NNFL. Eliminating correlations between variables can significantly simplify the assignation (identification) problem of the unknown and can help rationalize the parameters, which have to be included in the NNFL or have to be measured from the unknown material.

There are several multivariate statistical methods available to use for the identification of an unknown material, such as clustering systems, principal component analysis, factor analysis or discriminant analysis.[4] Each technique could be used for nuclear forensics purposes, however, for the Galaxy Serpent exercise, where the task is (i) to assess whether the hypothetical seizure is or is not consistent with the reactors assigned, and, (ii) to answer from which reactors the unknown sample may derive, possibly the best suitable approach is the linear discriminant analysis (LDA). This method is used for classifying observations into two or more groups, if we have already a library with known and pre-defined groups. LDA is a supervised classification (learning) technique, which uses a training (learning) data set to construct a model to differentiate the groups, which are the different reactors in our case now. The setup of the model is based on the variables present in the NNFL by establishing the discriminant function with the linear combination of independent variables (also called predictors) that creates the maximum difference (distance) between groups. The validation of the model is usually done with the classification matrix (confusion matrix) and cross-validation. Having built the model, one can also predict a group membership for an unknown sample using the discriminant function.

LDA Classification of NNFL Exercise Data

Minitab v.17 software (Minitab Inc., PA, USA) was used for the calculations. In order to eliminate correlating variables, missing values or non-equal number of data points for the variables, four variables (predictors) were used: ^{239}Pu/Pu$_{total}$, ^{240}Pu/Pu$_{total}$, ^{241}Pu/Pu$_{total}$ and ^{235}U/^{238}U. Note that initially most variables, which were complete in the present test exercise, were included in the training set (e.g., burnup values), but as the variables are often highly correlated, there is no need to increase the number of variables. Three Siarnaq samples contained missing values, thus they were left out of the learning set. It was assumed that the data of the variables represent a sample from a multi-variate normal distribution and the variance/covariance matrices of variables are homogeneous across groups. The summary of LDA classification as the classification table (confusion matrix) is shown in Table 1. The classification table is simply a table (matrix) in which the rows are the observed (calculated) cases of the training set based on the model, while the columns are the true (known) group assignations used to build the model. If the prediction is perfect, all cases will lie on the diagonal. The percentage of cases on the diagonal is the percentage of correct classifications. As shown in Table 1, the LDA correctly assigned fifty-five cases out of fifty-seven (96.5 percent correct), although the probability of correctly classifying the Anthe and Hyperion group members are somewhat worse (88.9 percent and 91.7 percent, respectively), while all Siarnaq cases were correctly assigned.

It has to be noted that the classification matrix often overestimates the correctness of the classification compared to the true error rate. This is because the data being classified are the same data used to build the classification function. The cross-validation routine works by omitting each observation one at a time, re-calculating the classification function using the remaining data, and then classifying the omitted observation. This

Table 1. Summary of classification (confusion matrix)

Anthe		True group		
		Hyperion	Siarnaq	
Assigned to	Anthe	8	0	0
	Hyperion	1	11	0
	Siarnaq	0	1	36
Total number		9	12	36
Correct (number)		8	11	36
Correct (proportion)		88.9%	91.7%	100%

Table 2. Summary of classification accuracy using cross-validation

Anthe		True group		
		Hyperion	Siarnaq	
Assigned to	Anthe	7	0	0
	Hyperion	2	11	0
	Siarnaq	0	1	36
Total number		9	12	36
Correct (number)		7	11	36
Correct (proportion)		77.8%	91.7%	100%

Table 3. Squared distances between groups

	Anthe	Hyperion	Siarnaq
Anthe	0.00	16.91	63.32
Hyperion	16.91	0.00	20.65
Siarnaq	63.32	20.65	0.00

Table 4. LDA prediction for the unknown Terpsichore samples

Unknown sample	Predicted group	Squared distance from group		Probability
Terpsichore-1	Siarnaq	Anthe	75.0	0.000
		Hyperion	37.4	0.000
		Siarnaq	7.5	1.000
Terpsichore-2	Siarnaq	Anthe	82.1	0.000
		Hyperion	45.4	0.000
		Siarnaq	11.9	1.000
Terpsichore-3	Siarnaq	Anthe	64.4	0.000
		Hyperion	23.3	0.000
		Siarnaq	0.7	1.000
Terpsichore-4	Siarnaq	Anthe	67.9	0.000
		Hyperion	23.6	0.000
		Siarnaq	0.1	1.000

process is repeated with each case left out in turn. Therefore, cross-validation is a more honest presentation of accuracy than that provided by the original classification table. The summary of classification correctness using cross-validation is shown in Table 2.

Based on the cross-validation the overall correctness of classification (i.e. the goodness of the model) is 94.7 percent, less correct for the Anthe and Hyperion groups (77.8 percent and 91.7 percent), and all the Siarnaq samples were correctly classified. The predictive accuracy of the LDA model also highlights the requirements of data quality in the NNFL data set: the best prediction could be achieved for the Siarnaq, where the highest number of samples was included in the library (thirty-nine) in contrast to Anthe and Hyperion reactors (number of samples are nine and twelve, respectively). Therefore, the more comprehensive the NNFL is, the better the predictive capability of multivariate analysis.

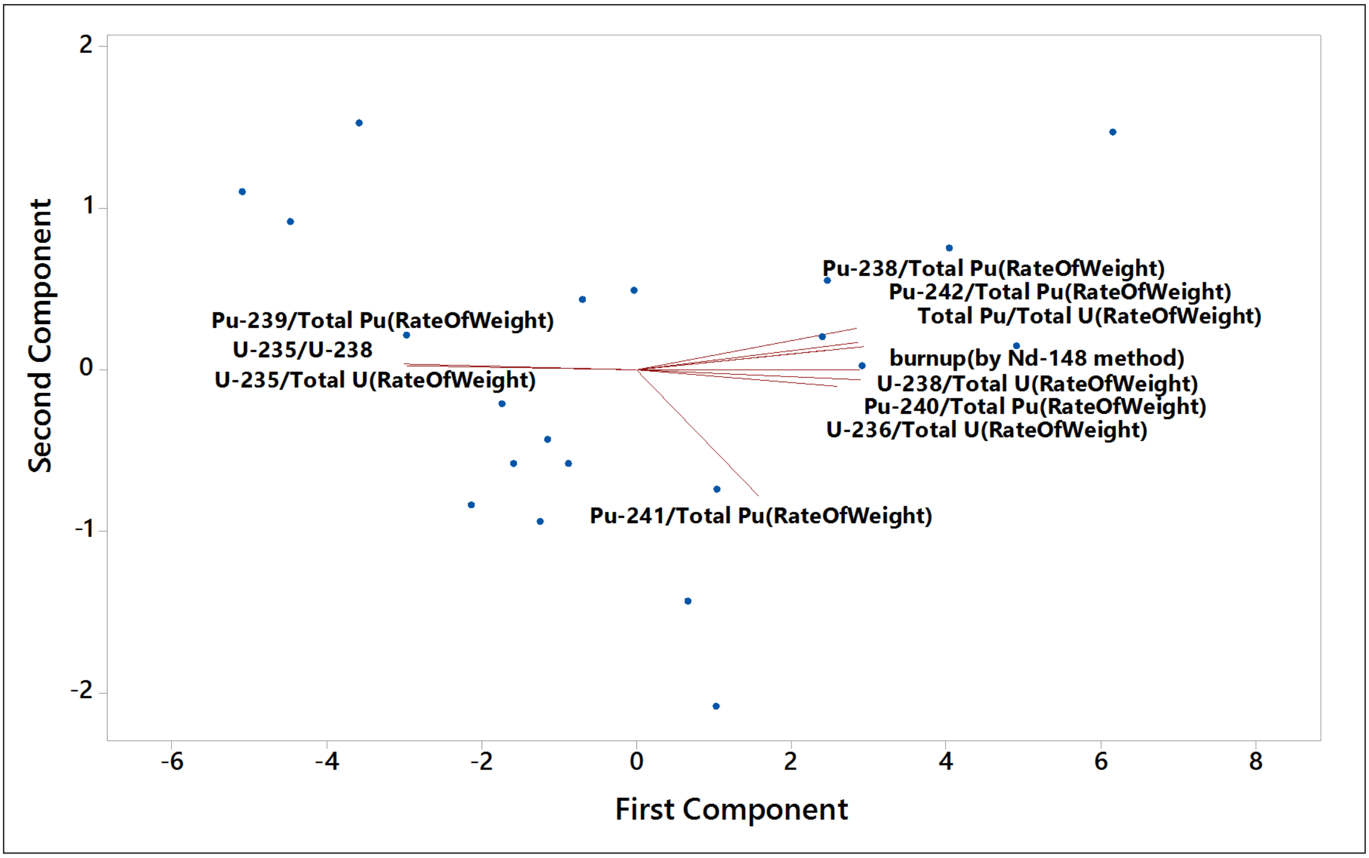
Using such calculations, not only is the linear discriminant function obtained, but also the group means of the predictor variables (also known as the centroids) and the squared distances between the groups (Table 3). The higher the distance between the groups, the better they are separated from each other. From Table 3 it can be seen that Siarnaq forms a very distinct group, well separated from the other two groups.

Predicting Group Membership of the Terpsichore Samples

Using the validated model, we can also predict the group membership of the four unknown Terpsichore samples. The measured nuclear data of the unknowns can be substituted into the calculated model discriminant function and the samples can be assigned to the group with highest function value. The assignation is based on the distance between the observed samples from the centroids of the groups of the learning set. The predicted group memberships of the Terpsichore samples are given in Table 4. Table 4 summarizes the predicted group for each Terpsichore subsample, their squared distances from the learning set centroids (i.e. the measure of similarity) and the calculated probability of prediction.

It can be seen from Table 4 that the low burnup Terpsichore samples (Terpsichore 1 and 2) have slightly higher distance from the Siarnaq learning set samples, but it is still significantly closer than to the other two groups, and the model predicts the assignation to the Siarnaq group with very high probability

Figure 4. Loading plot of the PCA analysis. The red lines are the original variables in the space of the first and second component.

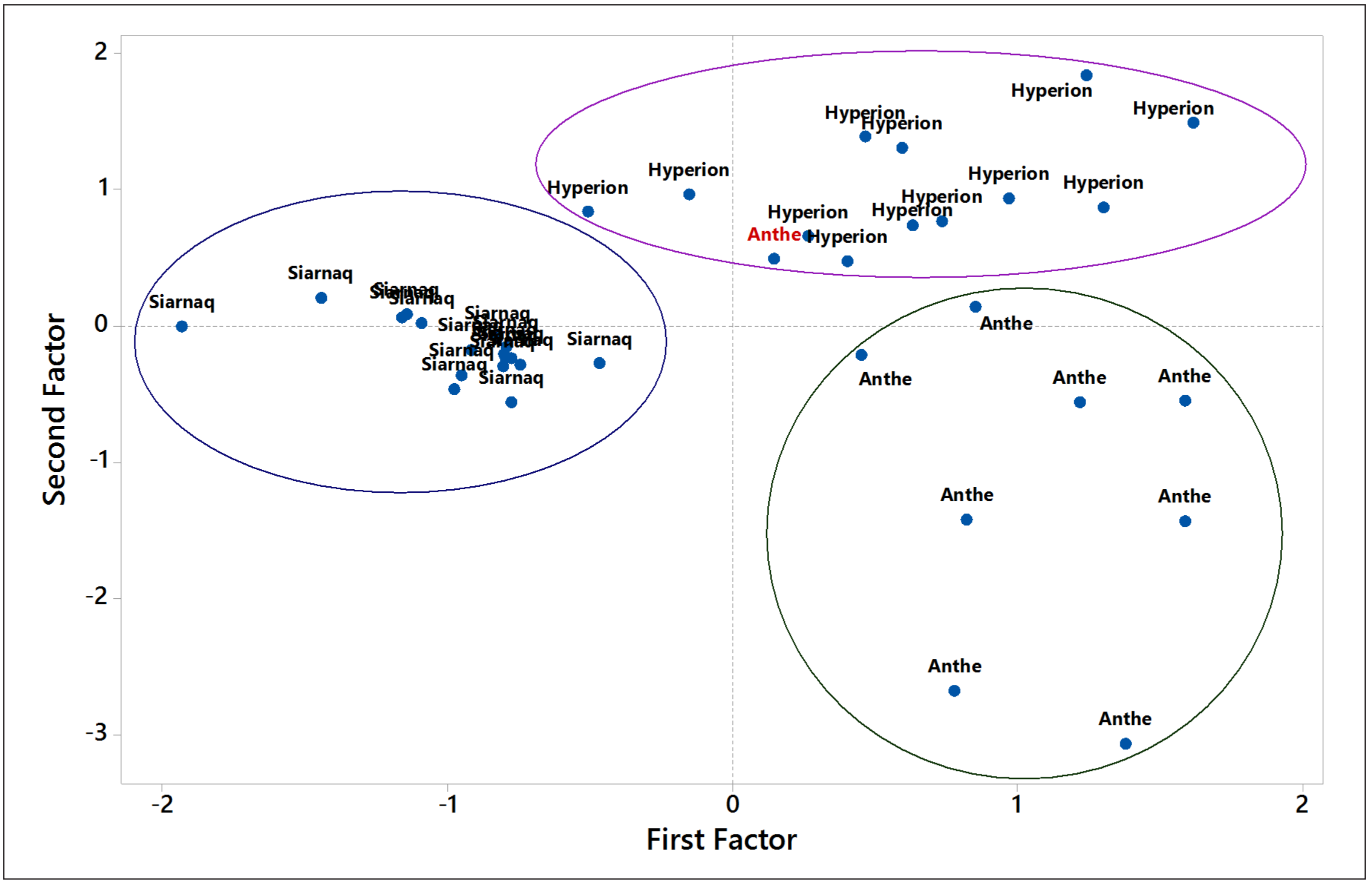


(above 99.9 percent). The LDA model in this case works also well in spite of extrapolation. The higher burnup Terpsichore samples (Terpsichore 3 and 4) are very close to the Siarnaq learning set centroids and are predicted to belong to the Siarnaq group with very high probability (above 99.9 percent). Note that for the LDA calculation, an assumption was made that the covariance matrices are equal for all groups. Calculations were also performed without this assumption using quadratic discriminant analysis, which gave the same prediction for the questioned samples with even better classification probability.

Other Uses of Multivariate Techniques

Other multivariate statistical techniques can also be effectively used if a NNFL is available. For instance, principal component analysis (PCA) can help identify the meaningful variables in the NNFL. PCA is a statistical procedure that uses orthogonal transformation to convert a set of observations of possibly correlated variables into a set of values of linearly uncorrelated variables called principal components. By the dimension reduction of the transformed data the internal structure of the data can be revealed, which best explains the variance in the data. This cannot only simplify the NNFL by eliminating redundant (correlating) variables, but it can also accelerate the identification by focusing on the relevant measurable parameters. Performing PCA on the Galaxy Serpent exercise, data several correlations could be identified using the loading plot (Figure 4). The loading plot is a plot of the relationship between the original variables projected onto the space of the principal components, and can be used to interpret relationships between the original variables. From Figure 4 it can be seen that several variables are highly correlated, thus the NNFL can be significantly simplified: the ratio of fissionable nuclides (^{235}U and ^{239}Pu) and the ^{235}U/^{238}U ratio are inversely proportional to the burnup, while the normalized values of ^{236}U, ^{238}Pu, ^{240}Pu, ^{242}Pu or the ^{238}U/U$_{total}$ are in correlation with the burnup of the spent fuel as expected for a nuclear reactor. The behavior of ^{241}Pu/Pu$_{total}$ is somewhat different, possibly due to the variable cooling times. By knowing the relationships between the variables, the content of the NNFL can be optimized without including redundant data.

Figure 5. Score plot of the FA analysis



A similar multivariate statistical technique, factor analysis (FA), can be used to describe variability among observed correlated variables to potentially lower the number of unobserved variables called "factors." Factor analysis searches for joint variations as a result of unobserved latent variables. The observed variables are modelled as linear combinations of the factors, including also error terms. In contrast to PCA, the emphasis in factor analysis is the identification of underlying factors that might explain the dimensions associated with large data variability. Similarly to PCA, plotting the data points projected onto space of the first two factors, which are usually responsible for the bulk of the variance, we can identify different clusters of the data (Figure 5). Note that the Siarnaq samples are distinctly separated from the other two reactors types even in the projection of the first two factors.

The above considerations have shown that multi-variate analysis can be used very effectively for the identification of an unknown spent fuel sample if comparison data (i.e., a "learning set") are available. A national nuclear forensics library may constitute such a resource of comparison data on samples of known origin and process history. Multivariate techniques can also cope with missing values, outlier unknowns or values with higher uncertainty at a certain degree, though the prediction probability will be significantly worse in such cases.

Conclusions

In the framework of the ITWG exercise Galaxy Serpent, a model national nuclear forensics library was built using a simple Excel spreadsheet. The library included data sets of spent fuel samples from three different reactors (Anthe, Hyperion, and Siarnaq). A data set of an unknown material (Terpsichore) was found to be consistent with the Siarnaq reactor. Using the methodology and terminology of the ITWG Guideline for Graded Nuclear Forensics Decision Framework the two materials are considered suggestive positive. This conclusion was obtained and corroborated, using two completely independent approaches. The first one is based on the isotope correlation technique and makes use of a simple and transparent two-dimensional visualization of the data. The second approach makes use of multivariate analysis, hence exploiting complex, multi-dimensional data sets.

The exercise also showed that selected techniques, such as principal component analysis or factor analysis, can be effectively used to optimize the NNFL content by eliminating redundant (correlating) variables. This knowledge may be used to reduce time and effort for both setting up a NNFL and identifying an unknown sample.

Maria Wallenius started her radiochemist career in Finland at the University of Helsinki in the nuclear safeguards project. In 1996, she moved to Institute for Transuranium Elements (ITU) to do research on characteristic parameters for the origin determination of plutonium in nuclear forensics, and she obtained her PhD in 2002. Since then she has been involved in various research and training activities in the nuclear forensics field. She is also the coordinator of the nuclear forensics analysis at ITU.

Zsolt Varga received his PhD in chemistry in 2007 from the Institute of Isotopes of the Hungarian Academy of Sciences. After working there as a research fellow until 2008, he joined the nuclear forensic group at the Institute for Transuranium Elements (ITU) of the European Commission Joint Research Centre first as a postdoctoral researcher, then as a scientific officer. His current research interests include the elemental and isotopic analysis of illicit nuclear materials and origin assessment of uranium samples of unknown origin.

Klaus Mayer has more than twenty years of experience in nuclear science and applications and is the author of more than 120 scientific publications in this field, including peer-reviewed articles, book chapters, and conference papers. He specializes in nuclear material analysis for safeguards purposes, and he has many interactions with IAEA and with Euratom Safeguards. In recent years, the analysis of nuclear material of unknown origin, also referred to as "nuclear forensics" has been in the focus of his research. From 1997 to 2010, he chaired the ESARDA Working Group on Destructive Analysis. Today, he is in charge of the activities on combating illicit trafficking and nuclear forensics at the European Commission's Joint Research Centre, Institute for Transuranium Elements (ITU). Since 2004, he has been the co-chair of the Nuclear Forensics International Technical Working Group (ITWG).

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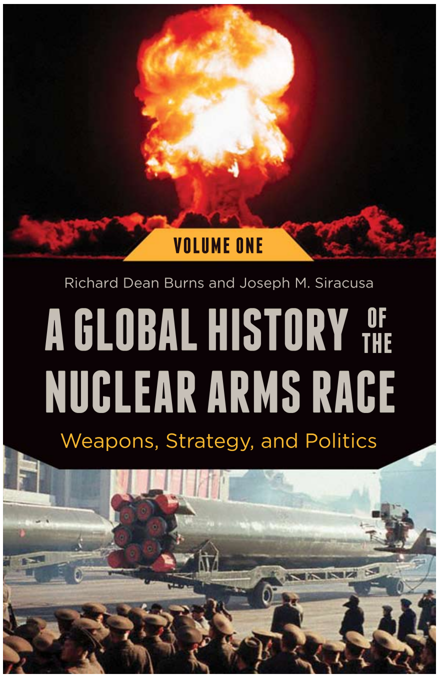
Book Review

By Mark L. Maiello,
Book Review Editor

A Global History of the Nuclear Arms Race

Weapons, Strategy, and Politics
Richard Dean Burns and
Joseph M. Siracusa
Hardcover, 642 pages in two volumes,
ISBN 978-1-4408-0094-8
Praeger Security International,
ABC-CLIO, LLC 2013



Fanfare is deserved for this effort by two masters of the history of nuclear weapons. This is an eminently readable, carefully crafted, and ultimately satisfying account of armaments, tactics, and policies from World War II to nearly the present day. Composed by professor emeritus and director of the Center for Armament and Disarmament at California State University (Burns) and by professor of human security and international diplomacy at the Royal Melbourne Institute of Technology (Siracusa) — both prolific authors — this presentation will be of great educational value to anyone and everyone entering the field of nuclear proliferation. It is a scholarly and I daresay an enjoyable means to an education about how the world's polities have inherited their nuclear weapons.

The book is divided into two volumes separating the discourse into the 1930s to 1980s timeframe (volume one); while volume two takes us to the present day. Volume one contains the entire work's table of contents whereas volume two carries the in-depth index and selected references for the collective effort. There are illustrations but not many and they are mostly of secondary importance to the well-conducted narrative. Several tables primarily useful for summarizing weapons specifications and a useful two-page "sidebar" on ballistic missile basics also buoy the text. These however are not the strengths of the volumes. The intellectual gratification lies squarely in the writing. Clear, rather concise, and occasionally hard-hitting when opinion is called for, the outstanding writing makes purchase of the book so very worthwhile.

The level of deep historical research is quite evident. The vaults of unclassified material appear to have been thrown open. Of particular value is the information about nations other than the United States: the writings and opinions of the Soviets for example on such matters as the arms race and the Cuban missile crisis. Such insight reveals the suppositions, assumptions, and the guesswork of politicians, their subordinates, and their intelligence agencies as they tried over the course of decades to maneuver their nations successfully over the nuclear chessboard. One finds alarmingly and frustratingly that the United States time and time again overestimated the nuclear capabilities of the Soviet Union (and was not very accurate about China as well).

As a student at Manhattan College, I had been taught that education is repetitive. Rarely does the subtlety of any material find understanding at first blush. The authors, for another reason entirely, made subtle use of this technique, which I think, proves invaluable to readers of this history. Creatively repeating the major events now and again obviates the need to flip back to previously covered ground when it may become unclear or forgotten. The authors have done this for us. Their reason is just as valuable: so that the book need not be read cover to cover. Chapters can stand alone. That works fine for research purposes, but the crisp thoughtful writing does permit the recommended cover-to-cover read.

Another approach the authors use honors the players in these affairs — the scientists, military leaders, politicians, and the scholars who study them — by quoting their assessments of the events. This is another reason for the healthy discourse found in this history and is especially rewarding when the players disagree. There is little analysis from the authors.

They flatly claim that drawing conclusions lies squarely with the reader. Their credentials and expertise surely gave them license to expound and occasionally they cannot resist. Their subtle critique of the George W. Bush administration's approach to treaties in general, the Comprehensive Test Ban Treaty in particular, and the fruitless search for weapons of mass destruction in Saddam Hussein's Iraq was factual and refreshing to encounter. The authors also reveal (and implicitly comment on) Henry Kissinger's and President Richard Nixon's unique and disparaging language of diplomacy especially with regard to India.

If the narrative bogs down at all, it is in the descriptions of weapons—both nuclear and those systems that can carry them. However, this comprises a relatively small portion of chapter 6. For my money, the politics and the international strategies are where the action lies. But to be fair, ballistic missile development simply cannot be ignored. Another "system" that cannot be discounted, Reagan's "Star Wars" initiative (officially called the Strategic Defense Initiative), had significant effects on the relations between the United States and the Soviet Union. Not as tangible and nor dry a subject as ballistic missiles, Star Wars receives a discussion by the authors commensurate with its impact. With that in mind, it is reasonable that previous and far more realistic and practical systems were discussed in volume one but just not with the same aplomb one encounters when the authors address the personalities and issues that shaped the post-atomic bomb world.

This minor glitch aside, the authors must be commended for their level of detail. Clearly they meant this work for serious students but whether deliberate or not, they found the point where I think, the general reader is also well accommodated. They did have one advantage: the history they convey is intrinsically interesting. Given that, the authors could still have sunk the ship with a boring treatise crushed under a weight of names, dates, and mere facts. But they hit the mark between scholar and interested reader; between need-to-know and nice-to-know. One wonders what heights this work could attain if the authors were as well known in popular circles as they are in academia — or if a strong campaign to commercialize the book was instituted. Nonetheless, Burns and Siracusa have achieved the brass ring of all technical writers: they verge on general popularity.

Details are where the devil lives but in these volumes, one can wade into them with confidence and courage. An example of the authors' ability to teach scholar and layman alike is their approach to the history of National Security Council Policy Paper Number 68 (NSC-68), that for all intents and purposes outlined U.S. nuclear strategy for decades since its inception on April, 7, 1950. Basically a grim assessment of the world view of that time (the Soviets were growing militarily), and obscure to most laymen, NCS-68 proffered possible courses of action that the United States could take to stem the influence of its communist rival. Long-serving government advisor Paul Nitze is introduced as its primary author and he along with government colleagues and scholars are quoted extensively throughout the subsequent discussion. The importance of NSC-68 and its legacy are nicely debated, providing a well-considered discourse on an important subject that eventually led to U.S. development of the hydrogen bomb. An important document is painlessly given an edifying treatment. Education is well served. The reader is not burdened.

The proliferation of nuclear arms to other powers such as the Soviet Union, Great Britain, China, India, and the rest is treated with the same dedication to scholarly research and readability. Stalin's doctrines and strategies, so crucial to an understanding of the early phases of nuclear proliferation, and Khrushchev's realignment of them, revealing that he too understood the dangers of nuclear warfare, are given excellent dissertations. How then the Cuban Missile Crisis? See Khrushchev's explanation in chapter 10 about the Soviet's desire to frighten the Americans (not to precipitate a war), and the Soviet confusion over the hostile American reaction. After all, did not the Americans have missiles staged in Turkey that were aimed at the Soviet Union? This is what objective history should strive for: the perceptions of both sides. Let the reader mull it over.

Burns and Siracusa also successfully tackle the question that spanned a few presidential administrations about the efficacy of anti-ballistic missile technology (it arose again with Star Wars), the Eisenhower years when the build up of nuclear arms ignited and the atoms for peace initiative sought to reign in proliferation, and the events more familiar to the current generation of non-proliferation experts: the birth of the International Atomic Energy Agency and the Nuclear Nonproliferation Treaty (NPT). Nuclear development in Israel, India, Pakistan, and North Vietnam, all receive treatment due their importance and impact on world events. The NPT and its evolution takes us to nearly the present day as the authors analyze the most recent NPT review sessions, the interactions of NPT participants in the post Cold War era and the initiatives complementing the treaty such as Nuclear Free Zones.

These two relatively slim volumes (eighteen chapters in all), fill what the authors describe as a lacuna — a gap — in the historical literature apparently because a world view of nuclear weapons development did not until now exist. If indeed they have filled such a void, they have done so in handsome fashion. Where it was possible, they have to an extent humanized it — certainly not by formulating biographies — nothing of the sort. But they did draw on enough of the personalities that made history to create an interesting essay. Here are Presidents Truman and Eisenhower battling the Soviets and the specter of runaway proliferation. Here is President Johnson deciding to live with the Chinese bomb. We also find Nixon and Brezhnev concluding the anti-ballistic missile treaty and Reagan and Gorbachev reaching out to each other in an attempt to end the arms race.

This work is a quiet treasure produced by modest professors who may or may not realize how close they have come to perfection.

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  1. Jones, F.T., and L. K. Chang. 1980. Article Title. *Journal* 47(No. 2): 112–118. 2. Jones, F.T. 1976. *Title of Book*, New York: McMillan Publishing.
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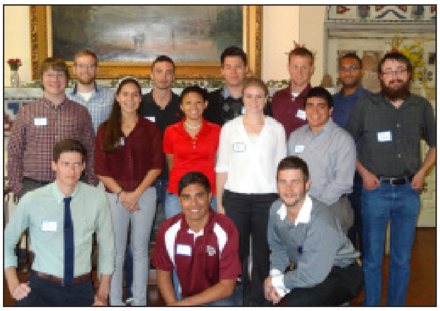
Throwing Down the Gauntlet to the Next Generation of Nuclear Stewards — the Enduring Nuclear Legacy

By Jack Jekowski
Taking the Long View Editor and Chair of the INMM Strategic Planning Committee

On April 14, 1998, at the first meeting of the newly formed Southwest Chapter of the INMM, held at the DOE Central Training Academy (now the DOE National Training Center) in Albuquerque, New Mexico, USA, then-President of the INMM Obie Amacker "threw down the gauntlet" to the attendees, challenging the new chapter to help the Institute engage the "next generation of nuclear stewards." At the time, the Institute's Executive Committee had been expressing growing concern with the aging of the membership and the lack of participation by the younger generation. They hoped to identify strategies to better engage students, universities and those entering the disciplines of nuclear materials management in the activities of the Institute.

Now, sixteen years later,[1] we are beginning to see the fruits of those efforts as our student chapters almost outnumber all of our U.S. and International Chapters combined, and attendance at our Annual Meeting is now more than 10 percent students, with many of them presenting technical and policy papers on critical issues.

The First Decade of the New Millennium



The student initiatives in the INMM began to take shape in the early part of the first decade. There were many supporters, including J. D. Williams,[2] John Matter, Dennis Mangan, Scott Vance, and others who picked up the gauntlet to ensure that the efforts became sustainable. This included changes to the bylaws to include special support for students, encouragement for the formation of student chapters, and the Annual Meeting Student Paper competition.[3] Texas A&M was the first student chapter authorized by the Institute in 2005. Their advisor, Dr. William Charlton, is still active today, bringing ten students to the recent SW Chapter's Annual Technical Meeting in Taos, New Mexico, USA, this May, with three students presenting technical papers (see adjacent photo of all of the students who attended this meeting, including a presenter from the University of New Mexico Student Chapter).[4] Today, the Institute has fourteen student chapters, including two international chapters,[5] and more than 150 student members (more than 10 percent of the total Institute membership). Two additional international student chapters are also going through the approval process, and may be added by the end of August.

Painting a Broad Picture of the Future — The Enduring Nuclear Legacy

The new millennium has proven to be far more challenging and threatening than many anticipated during the economic boom era of the 1990s. Starting with the nuclear tests by India and Pakistan in 1998, and amplified by the tragic events of September 11, 2001, and the subsequent decade-long conflicts in Iraq and Afghanistan, the continuing issues with the nuclear programs in Iran and North Korea, and the repercussions of the Fukushima nuclear accident, these challenges cover a broad spectrum of geopolitical and scientific/technical topics that create a difficult and, at times, overwhelmingly complex environment.[6]

In recent years, these challenges have been detailed in a number of venues including:

- President Obama's April 5, 2009, "Prague Speech" in which he set the tone for his administration to pursue a goal of a world without nuclear weapons.[7] Also included in this major policy speech were the goals to negotiate a new verifiable Strategic Arms Reduction Treaty with Russia (New START, ratified in 2010); pursuit of U.S. ratification of the Comprehensive Test Ban Treaty (CTBT); pursuit of an international agreement on a Fissile Material Cutoff Treaty (FMCT); strengthening of International Atomic Energy Agency (IAEA) inspections; creation of an international nuclear fuel bank; and securing vulnerable nuclear materials around the world. These challenges and others were subsequently reiterated in the U.S. National Security Strategy,[8] and related documents, including the Quadrennial Defense Review[9] (2010 and 2014), and the Nuclear Posture Review.[10] After five years of efforts, however, much still needs to be done to achieve some of these goals.

- President Obama's Nuclear Security Summits (2010, 2012, and 2014)[11] that have encouraged an extraordinary gathering of nations to identify and address the critical nuclear issues facing the world today.
- President Obama's "Brandenburg Gate Speech" on June 19, 2013, in which he reiterated many of the tenets of the Prague Speech, and suggested further reductions in the stockpiles of Russia and the United States by one-third would be possible.[12]
- Various non-governmental organizations (NGOs), including the Nuclear Security Project,[13] the Arms Control Association,[14] the Federation of American Scientists,[15] the Nuclear Threat Initiative,[16] the Bulletin of Atomic Scientists,[17] Global Zero,[18] and others have examined the critical issues associated with "things nuclear" and identified recommendations and actions for the international community to take.[19]

Also, in previous columns, I have posed a number of challenges/questions to the membership of the Institute to stimulate strategic discussions.[20]

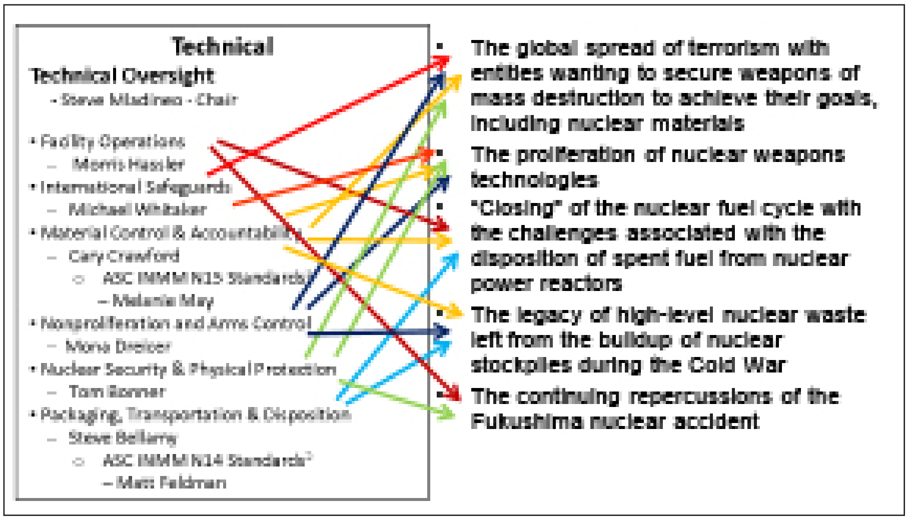
Within the context of all of these perspectives, I assembled five "Great Challenges" to be "thrown down" to the next generation of nuclear stewards and presented them to the SW Chapter's Annual Technical Meeting in May. These Great Challenges have subsequently been vetted with a small group of senior members in the Institute and modified to reflect initial feedback, including some significant recommendations from Jim Larrimore, former chair of the INMM International Safeguards Technical Division. This is our ***enduring nuclear legacy***:

- Keeping nuclear materials around the globe secure and out of the hands of entities wishing to have access to them for nefarious purposes
- Controlling the spread of dual-use nuclear technologies, especially uranium enrichment and plutonium reprocessing
- Completing the "back-end" of the nuclear fuel cycle with the safe and secure handling and disposition of spent fuel from nuclear power reactors
- The legacy of high-level nuclear waste left from the buildup of nuclear stockpiles during the Cold War
- Developing nuclear power that is proliferation-resistant and accepted by the general public as safe, economically viable and environmentally friendly

These five challenges capture much of what has been voiced in different venues and provides a snapshot into the difficult world ahead that our younger generation is inheriting. These five challenges are presented here to stimulate strategic discussions among the Institute's membership with the understanding that they can be reshaped to reflect the diversity of perspectives that constitute the collective knowledge of the Institute's membership.

Alignment with the INMM Mission and Technical Divisions

The five Great Challenges align well with the Institute's mission[21] and the Technical Division structure.[22] In fact, in an attempt to map the Technical Divisions into these five challenges, shown here, the chart quickly became unreadable since almost every challenge has a commonality with every Division. A recent presentation given by the author to the Southwest Chapter's Annual Technical Meeting elaborated on some of the component elements of each of these legacy Challenges.[23] All five of these Challenges included the element "Workforce Development — preparing the next generation



of nuclear stewards." Within the context of the Great Challenges, each nation has their own unique issues to address as well.[24]

Throwing Down the Gauntlet to the Next Generation

Just as Obie Amacker threw down the gauntlet to the Southwest Chapter in 1998, now the time has come for the INMM to throw down the gauntlet to that generation. The enduring nuclear legacy is a challenge to the very future of mankind, and it is up to us to ensure that we have done all we can to enable the next generation to pick up that gauntlet and accept the responsibility.

The INMM is continuing to enhance the participation of the next generation through a number of actions:

- Strengthening student and university engagement through senior management oversight on the Education and Training Committee[25]
- Encouraging student participation in the INMM Annual Meeting, and in the affairs of the institute by including representatives "at the table" in our Executive Committee meetings
- Broadening international membership participation, including encouraging the formation of international student chapters
- Creating the enhanced organizational structure three years ago to address the "Great Challenges" of the 21st century
- Examining the broader strategic role for the Institute in standards development and collaborations
- Promoting the importance of the continuing role for the Institute internationally as well as in the United States

It is critical that all of our membership recognize the importance of supporting, mentoring, and encouraging the next generation to ensure that the enduring nuclear legacy is safely sustained long into the future. Those of you who attend the INMM Annual Meeting can certainly seek out our student members and welcome them, as well as volunteer to help mentor those students during the meeting. Others can use their spheres of influence within their chapters to offer assistance to a local student chapter, whether it is through guest lectures, encouragement to attend chapter meetings and the Annual Meeting, or staying in close touch with student chapter advisors who bear the burden of sustaining those organizations as students matriculate.

This column is intended to serve as a forum to present and discuss current strategic issues impacting the Institute of Nuclear Materials Management in the furtherance of its mission. The views expressed by the author are not necessarily endorsed by the Institute, but are intended to stimulate and encourage JNMM *readers to actively participate in strategic discussions. Please provide your thoughts and ideas to the Institute's leadership on these and other issues of importance. With your feedback we hope to create an environment of open dialogue, addressing the critical uncertainties that lie ahead for the world, and identify the possible paths to the future based on those uncertainties that can be influenced by the Institute. Jack Jekowski can be contacted at jpjekowski@aol.com.*

Endnotes

1. See Jekowski, J. 2002. Preparing the Next Generation Nuclear Stewards. *Journal of Nuclear Materials Management*, Volume 30, No. 2, pp. 10-15, for an historical perspective on the first three years of efforts by the Southwest Chapter and the Institute to launch the next generation initiative. The INMM President's Message in that same edition of the *JNMM*, "The Graying Nuclear Workforce" by J. D. Williams, and the Technical Editor's Note by Dennis Mangan also provide additional insight into these efforts.
2. J. D. Wiliams, who was an early champion of many of today's student initiatives, has posthumously been recognized for his efforts through the annual "J. D. Williams Student Paper Award" at the annual meeting.
3. Glenda Ackerman is the chair of the Student Paper and Scholarship Subcommittee, which oversees the annual J. D. Williams Student Paper Award competition. Ackerman is always seeking more help for paper reviewers and oral presentation evaluators at the INMM Annual Meeting as the number of eligible student papers have increased dramatically in the recent past.
4. Dr. Charlton was recognized by the Institute for his efforts over the years with a Special Service Award. Similarly, Dr. Sara Pozzi, the advisor for the University of Michigan Student Chapter, was recognized with a Meritorious Service Award two years ago for her amazing efforts to mentor and encourage students to participate in Institute activities,

including consistently sponsoring several students to attend the INMM Annual Meeting.

5. Willem Janssens is the Chapter Relations Committee chair who works with all of the INMM chapters to encourage their engagement in Institute activities. He has been working with Steve Ward, chair of the Education and Training Committee to develop more International student chapters.
6. See "As the World Turns" section in Jekowski, J. 2014. Reflecting on the Health of the INMM. *Journal of Nuclear Materials Management*, Volume 52, No. 3, for the most recent update to the global environment that the INMM must work within.
7. See http://www.whitehouse.gov/the_press_office/Remarks-By-President-Barack-Obama-In-Prague-As-Delivered
8. See http://www.whitehouse.gov/sites/default/files/rss_viewer/national_security_strategy.pdf
9. See http://www.defense.gov/home/features/2014/0314_sdr/qdr.aspx
10. See http://www.defense.gov/npr/
11. See http://www.state.gov/t/isn/nuclearsecuritysummit/ and https://www.nss2014.com/en
12. See http://www.whitehouse.gov/the-press-office/2013/06/19/fact-sheet-nuclear-weapons-employment-strategy-united-states and http://www.defense.gov/pubs/reporttoCongressonUSNuclearEmploymentStrategy_Section491.pdf
13. See http://www.nuclearsecurityproject.org/vision-and-steps
14. See https://www.armscontrol.org/
15. See http://www.fas.org/
16. See http://www.nti.org/
17. See http://thebulletin.org/
18. See http://www.globalzero.org/
19. Note: This is not an all-inclusive list of NGOs engaged in examining the future of the nuclear world. The Strategic Planning Committee, along with the help of many others, has compiled a matrix of organizations with nuclear-related missions, some of which align with the INMM. This list currently comprises more than fifty international organizations. This effort was designed to help the Institute identify possible collaborations.
20. Jekowski, J. 2012. Looking Back at a Decade of Tumult — and Looking Forward to an Uncertain Future. *Journal of Nuclear Materials Management,* Volume 40, No. 3, pp. 99-101.
21. Ibid.
22. For the most current functional organization chart, visit INMM's website, www.inmm.org/AM/Template.cfm?Section=Organizational_Chart1&Template=/CM/ContentDisplay.cfm&ContentID=4756.
23. See www.itpnm.com and click on "What's New" to view the May 2014 entry on the INMM SW Chapter Annual Technical Meeting.
24. See, for example, the continuing struggles here in the U.S. with the organizational structure of the National Nuclear Security Administration and the difficulties the U.S. faces in sustaining a viable Nuclear Security Enterprise: http://www.armed-services.senate.gov/hearings/14-04-09-national-nuclear-security-administration-management-of-its-national-security-laboratories-and-the-status-of-the-nuclear-security-enterprise
25. Steve Ward is the chair of the INMM Education and Training Committee, having taken over that role from long-serving chair, Debbie Dickman. Ward is also currently the acting chair of the Student Activities Subcommittee, having taken over that position from long-serving Dr. Mark Leek.

Calendar

August 24–28, 2014
8th International Conference on Isotopes and Expo
Hyatt Regency Hotel
Chicago, Illinois USA

September 17–18, 2014
WINS Best Practice Workshop on End of Life Management of Radioactive Sources
Novotel Paris Gare
Montparnasse Hotel
Paris, France

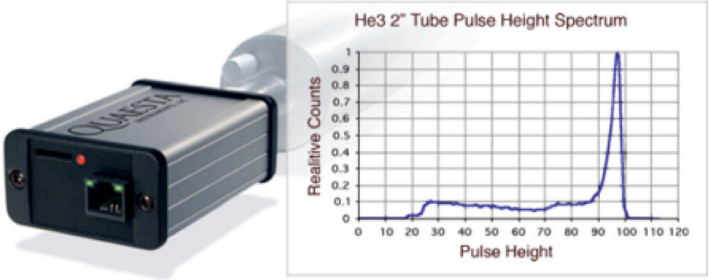
September 29–October 1, 2014
WINS Best Practice Workshop on Understanding and Mitigating the Insider Threat
ARES Tower
Vienna, Austria
Email: jadallah.hammal@wins.org for more information.

October 20–24, 2014
Symposium on International Safeguards: Linking Strategy, Implementation and People
Vienna, Austria
Organized by the International Atomic Energy Agency (IAEA) in cooperation with the European Safeguards Research and Development Association (ESARDA) and the Institute of Nuclear Materials Management

January 12–14, 2015
INMM 30th Spent Fuel Seminar
Crystal Gateway Marriott Hotel
Arlington, Virginia US A

July 12–16, 2015
INMM 56th Annual Meeting
Renaissance Esmeralda
Indian Wells, California USA

September 11–16, 2016
PATRAM 2016
Kobe Portopia Hotel
Kobe, Japan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INMM
INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT

Institute of Nuclear Materials Management

56th Annual Meeting

Meeting Program

July 12 – 16, 2015
Renaissance Esmeralda
Indian Wells, California USA

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