

An overview of recent and planned advances in the evaluation of environmental sampling and nuclear material characterization analysis results

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Abstract:

The competent and timely evaluation of environmental sampling and nuclear material characterization analysis results within the Department of Safeguards at the International Atomic Energy Agency (IAEA) is vital to providing assurances regarding the absence of undeclared nuclear materials and/or activities in a State and an important contribution to drawing credible safeguards conclusions. While analytical techniques and methodologies have continued to evolve over the years, the suite of software tools used to evaluate the analysis results has, until recently, remained mostly unchanged. In preparation for a potential significant increase in the number of environmental and nuclear material samples collected in the field and to leverage the multiple types of data being reported by the Network of Analytical Laboratories (NWAL), efforts have been undertaken to improve the tools available to IAEA evaluators in order to improve both the effectiveness and efficiency of evaluating the analytical data. This paper provides an overview of the recent advances in the software and data analysis techniques used by IAEA evaluators, as well as those envisioned for the near-future.

1. Introduction

In the 25 years since environmental sampling (ES) [1] was implemented as a safeguards verification tool, it has proven to be one of the most important tools available to the IAEA for detecting undeclared nuclear materials and/or activities at both declared and undeclared locations. Additionally, material characterization analysis of bulk nuclear material (NM) samples collected from various stages of the nuclear fuel cycle (NFC) allows the IAEA to verify the NM and processes declared at the NFC facilities.

As the ES program has expanded over the years, the analytical techniques and methodologies applied to environmental and nuclear material samples by the Network of Analytical Laboratories (NWAL) have continued to evolve and improve. These advancements have not only resulted in higher quality data (e.g. the introduction of the modified total evaporation method for multi-collector thermal ionization mass spectrometry [2]), but have also provided additional types of data (e.g. uranium particle age-dating [3, 4]).

IAEA evaluators are responsible for receiving and evaluating the multiple types of analytical data reported by the NWAL and comparing it to, where applicable, the nuclear materials and activities declared at the sampled location, the past sampling results related to the sampled location, and to models of the relevant NFC processes (e.g. uranium enrichment and irradiation) [5]. The inferences made in these evaluations regarding the consistency or inconsistency of the findings with the declarations are an important contribution to the safeguards conclusions drawn for a State. Therefore, it is essential that such evaluations are performed in a competent and timely manner.

In order to make the best use of the multiple types of data reported by the NWAL and in preparation for a potential significant increase in the number of samples collected in the field (e.g. if new safeguards agreements are concluded between the IAEA and Member States), IAEA evaluators have identified ways in recent years to improve both the effectiveness and efficiency of ES and material characterization evaluations, which includes the development and deployment of new software tools and improvements to those already used by IAEA evaluators. These tools originate from commercially available off-the-shelf (COTS) software, internally developed software, software provided to the IAEA through Member State Support Program (MSSP) tasks and in-kind contributions, and software developed by contractors.

This paper briefly describes tools that are currently in use, tools that are still being developed, and projects that are planned for the near future.

2. Commercially available off-the-shelf software

2.1. Microsoft Power BI

Recently, the Department of Safeguards (SG) has made Microsoft Power BI Desktop and Report Server available to SG staff, which allows for the development and internal sharing of visualization-rich dashboards. As Power BI has the ability to connect directly to a variety of different data sources (e.g. the Oracle-based ES database (ESDB)), dashboards have been developed to assist IAEA evaluators with many aspects of their work, including monitoring workload, monitoring sample status at the NWAL and monitoring evaluation timeliness. Figure 1 shows one of the dashboards used by IAEA evaluators to monitor their current and upcoming workload to ensure the timely delivery of evaluation reports to the Divisions of Operations.

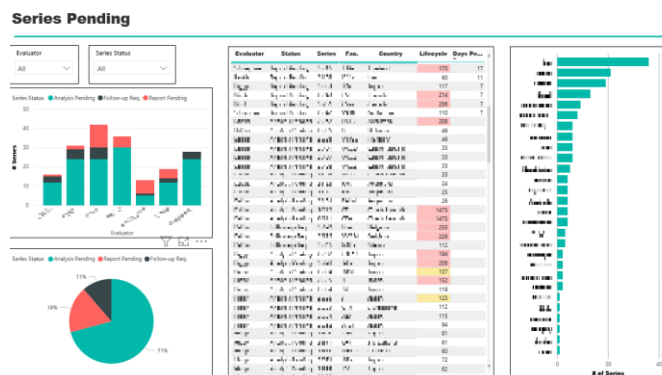


Figure 1. Power BI dashboard used by IAEA evaluators to monitor their current and upcoming workload

3. Internally developed software

In the course of their work, IAEA evaluators strive to identify new and improved methods for data analysis and visualization, as well as ways to streamline the data handling and reporting processes. Using various programming languages (e.g. Python, R, and JavaScript), several software tools have been developed (or are currently under development) that, for example, (i) streamline the querying, plotting, and exporting of relevant data, (ii) provide a way to integrate multiple types of analytical data (i.e. U/Pu particle isotopics and elemental concentrations) in an effort to identify NFC processes they may have resulted from, and (iii) allow for more effective comparisons of elemental compositions and morphological characteristics between particles.

3.1 ENVISDA

Until recently, the process of querying and plotting data, formatting plots, and creating evaluation reports has been cumbersome and required multiple steps. While other software tools have been introduced over the years (e.g. Cluster [6] and Visual ES [7]) to facilitate these processes, they have other limitations (e.g. no direct connection to the ESDB). However, both Visual ES and Cluster provide evaluation capabilities that are unique; Visual ES is primarily a tool which provides geospatial ES evaluation capabilities at the site, building, and room level, while Cluster is a plotting tool which allows for statistical analysis, such as cluster identification.

Environmental Sampling Data Analysis (ENVISDA) is a software tool written in Python that streamlines and modernizes all aspects of the ES evaluation process by providing, for example, (i) a direct connection to the ESDB, (ii) interactive plotting of current and historical data, (iii) the ability to import NFC modelling scenarios and calculate material mixing scenarios, (iv) easy access to relevant information (e.g. historical data related to the sampled locations, working papers, old reports), and (v) an evaluation report generator. Additionally, in certain instances when Microsoft Excel may be better suited for an evaluation, ENVISDA has a feature that automatically exports all queried data into a formatted Excel workbook, including automatic generation of plots. By automating these aspects of the ES evaluation process, ENVISDA has proven to save evaluators a significant amount of time when preparing reports. Figure 2 shows a screenshot of the graphical user interface (GUI) of ENVISDA that highlights a few of the features described above.

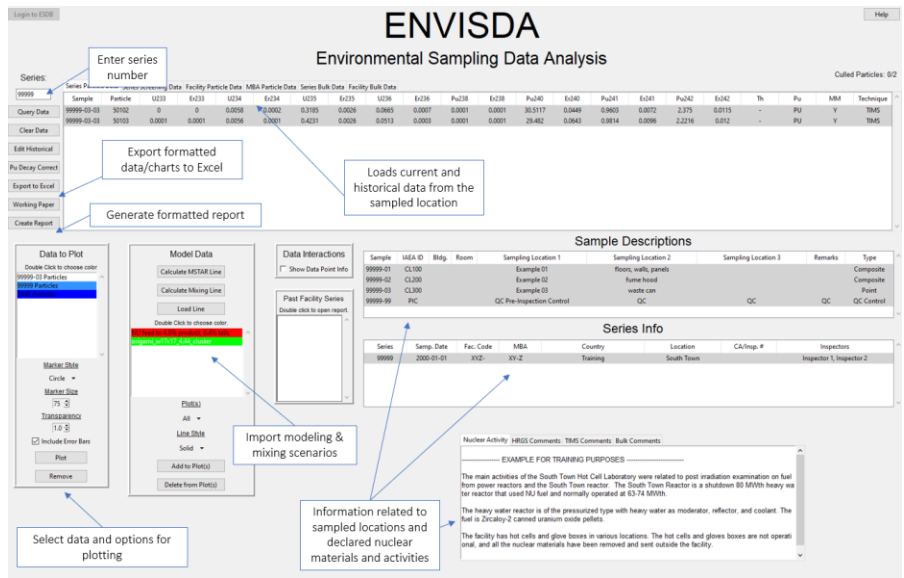


Figure 2. Screenshot of the ENVISDA GUI highlighting its features

3.2 Visual SEM

IAEA evaluators have been issuing an increasing number of analytical requests for elemental composition, morphology, and compound identification of particulate collected on swipe samples, which can provide additional assurances regarding the absence of undeclared nuclear materials and/or activities at the sampled location. As such, efforts have been undertaken at the IAEA to establish a particle library that catalogues the elemental and morphological data provided by the NWAL and to develop a software (i.e. Visual SEM) that allows IAEA evaluators to more effectively and efficiently investigate these data [8].

Visual SEM is written in JavaScript and links sample information, analytical results (e.g. elemental composition, particle size), and morphological images of particles and provides the ability for evaluators to compare these characteristics to those of particles from known materials and/or from particles on swipe samples collected at various NFC facilities. The software also allows for evaluators to search for particles within the library based on their elemental/morphological characteristics or their associated NFC activity (see Figure 3). Importantly, the software also allows evaluators to efficiently create ready-to-use figures for evaluation reports that combine the morphological images of one or more particles of interest, as well as the elemental chart that summarizes the elemental composition. The development of this library and software provides a significant improvement to the evaluation of elemental and morphological data at the IAEA.

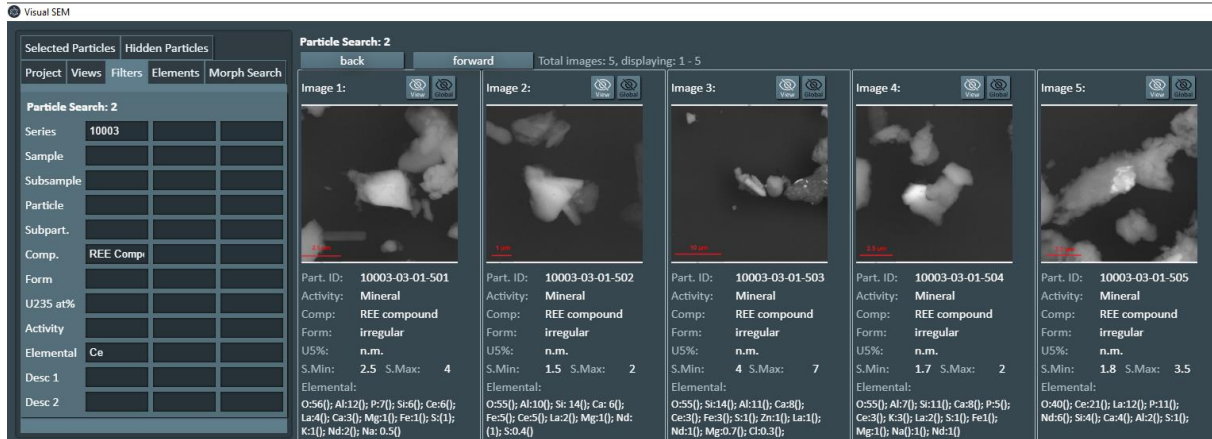


Figure 3. Particle search feature with Visual SEM [8]

3.3 LLAMA

The Linked Library for Material Analysis (LLAMA) is a software tool being written in Python that compares the materials generated/used throughout the NFC with analytical results from particles found on environmental samples. Using the measured elemental composition of a particle, a vector is created for comparison to a library of compounds. By minimizing the Euclidean distance between the particle composition vector and the vectors within the compound library, the compounds which are most similar to the measured particle can be identified. Using these most similar compounds, the associated NFC process can be extracted from the library and overlaid with the particle's uranium and plutonium isotopic signature to create a visual representation of the particles found on a swipe and their associated NFC process. Figure 4 shows an example of the fused data with particles identified as being indicative of reprocessing and enrichment activities.

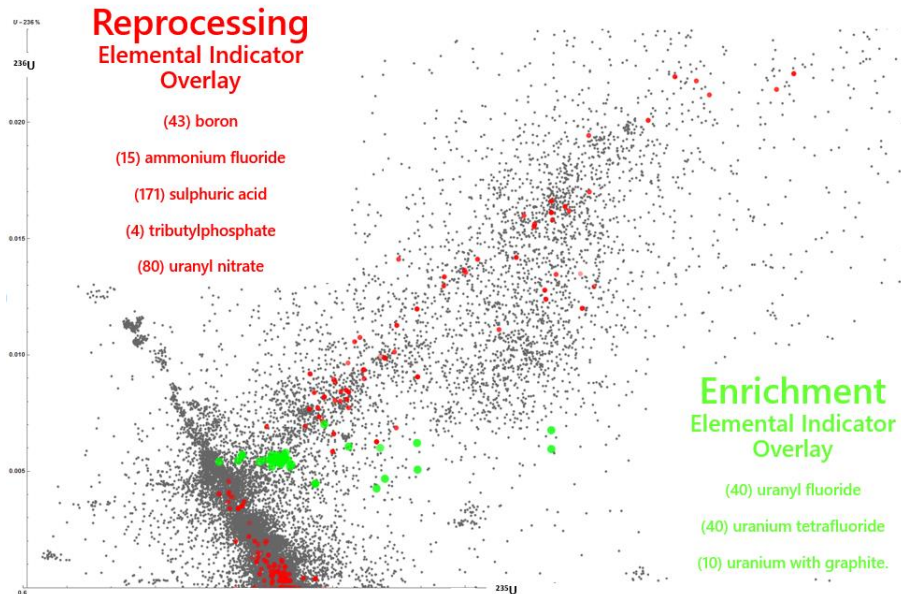


Figure 4. Data fusion within LLAMA identifying particles as being indicative of reprocessing and enrichment activities

3.4 OR2EX

ORIGAMI to Excel (OR2EX) is a software tool written in both Python and R that automates the use of the ORIGAMI reactor code [9] and the associated post-processing of the output files into a format that is suitable for use with the other tools used by IAEA evaluators (e.g. ENVISDA and Cluster). ORIGAMI is used to perform activation, depletion, and decay calculations using pre-generated ORIGEN libraries that are distributed with the SCALE code system. OR2EX implements the simplest version of the ORIGAMI code, where the fuel assembly materials are represented as a single mass lump (i.e. axial and radial power distributions are not considered).

While running ORIGAMI via the SCALE GUI (Fulcrum) is relatively straightforward, extracting the desired information and converting it into a format that is suitable for use in other software is less straightforward; automating this process makes reactor modelling more efficient. After selecting the reactor library of interest and inputting the desired parameters within the R Shiny GUI (see Figure 5), the code creates the SCALE input file, runs ORIGAMI, and post-processes the five output files via the Python script into the format appropriate for use with other software used by evaluators, where the modelled scenario can be compared to the measured data. Additionally, all relevant files are saved in an automatically generated, timestamped folder so that they can be easily referenced and/or reused when needed.

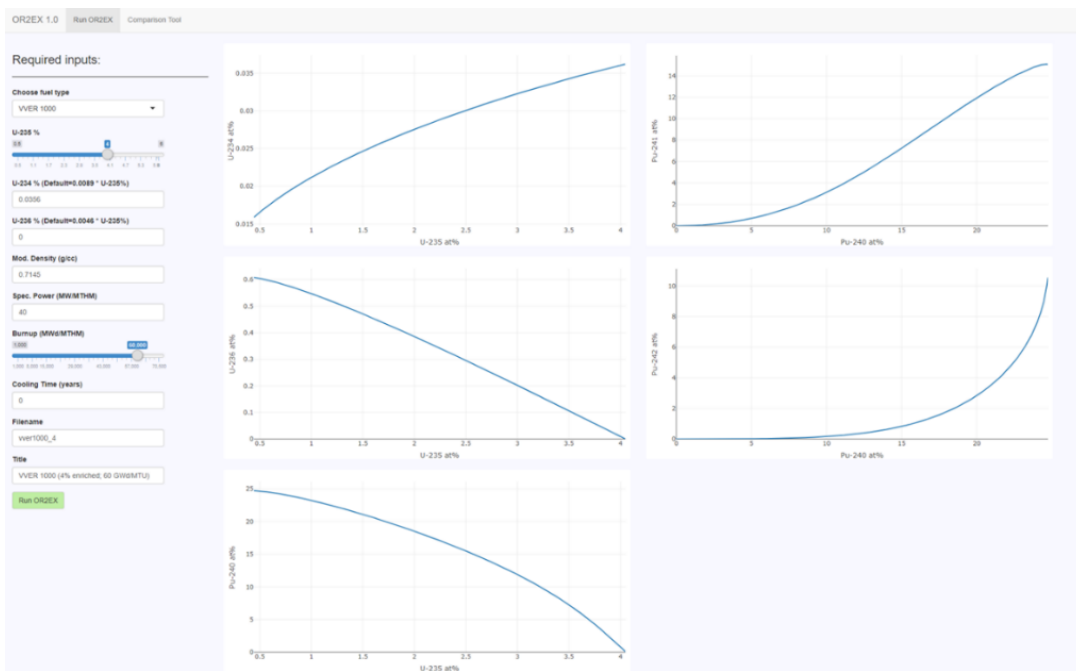


Figure 5. The OR2EX GUI developed in R Shiny showing the input parameters and the uranium and plutonium isotopic plots

4. Software provided through MSSP Tasks and in-kind contributions

Several tools have also been provided (or are currently under development) through Member State Support Program (MSSP) Tasks and in-kind contributions that aim to assist IAEA evaluators through NFC modelling, assessing the origin of uranium materials, and identifying outliers and clusters in particle datasets.

4.1 MSTAR2019

The collection, analysis, and evaluation of results from ES swipes/swabs and UF₆ samples from uranium enrichment facilities provides the IAEA with assurances that undeclared enrichment activities are not taking place. For example, evaluators look for certain indicators, such as (i) uranium enriched to higher than declared levels, (ii) the presence of undeclared feed materials, and (iii) the presence of an undeclared side product and/or feed.

When evaluating results from samples collected at uranium enrichment facilities, IAEA evaluators will often compare the analytical results with cascade models generated using the MSTAR code developed by Oak Ridge National Laboratory and provided to the IAEA as deliverable of the U.S. Support Program (USSP). The original MSTAR code was given to the IAEA in 1996, followed by an update in 2012. Recently, the USSP has delivered to the IAEA the latest version of MSTAR (MSTAR2019) that provides, among other things, (i) an improved GUI, (ii) the ability to model side product/feed streams, (iii) formatted outputs according to IAEA specifications, and (iv) the ability to run the executable via the command line that allows evaluators to run MSTAR through other software applications (e.g. ENVISDA) [10]. Figure 6 shows the GUI of MSTAR2019 and an example of how models generated are used to compare to analytical results from samples of UF₆ collected in the field.

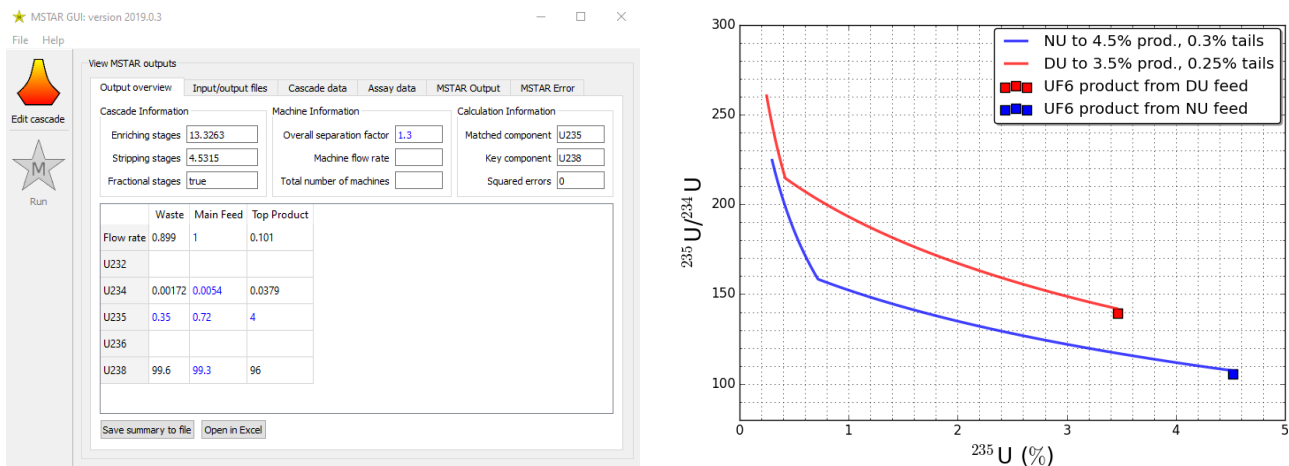


Figure 6. The MSTAR2019 GUI (*left side*) and an example of how MSTAR is used for comparing to measured data and declared information (*right side*)

4.2 Spotlight

Swipe and swab samples collected from uranium enrichment facilities are typically analyzed by particle analysis techniques, which determines the uranium isotopic composition in individual particles. Such techniques are useful for identifying multiple materials that may be present on a swipe (e.g. feed, product, and tails). IAEA evaluators must then determine if the particles detected on a sample indicate deviations from the declared NM and activities and/or the known/expected signatures of the facility. Currently, this is done by comparing the new findings on a sample to the historical signatures of the facility and the appropriate cascade models (e.g. from MSTAR) based on the declared NM and activities.

Spotlight is a software application being developed by Lawrence Livermore National Laboratory (LLNL) through a USSP Task [11] that will provide IAEA evaluators with robust, statistically-based tools to identify outliers (i.e. those findings significantly different from historical findings and/or modeled scenarios) and particle clusters (i.e. groups of similar particles that may indicate distinct feed, product, or tails materials).

The Spotlight GUI (being developed in R Shiny) will provide the necessary capabilities for IAEA evaluators to investigate the findings (e.g. interactive plotting, data culling, particle labelling) and prepare the appropriate outputs (e.g. plots, data summary tables) for reporting to the Divisions of Operations (see Figure 7).



Figure 7. The current status of the Spotlight GUI developed in R Shiny showing the results of one of the outlier detection engines and the ability to label particles

4.3 INDEPTH-ES

Forward depletion calculations (e.g. as implemented in OR2EX described above) require certain parameters (i.e. reactor type, initial isotopic composition of the fuel, specific power, irradiation time, cooling time) from the user to obtain the isotopic composition of the irradiated material. Such calculations are useful to evaluators when the most likely irradiation scenario is known (e.g. when evaluating a sample collected from a hot cell facility that performs post-irradiation examination on a single type of irradiated material). In other instances, however, a sample may be collected from a facility that has processed many different types of irradiated materials. In such cases, evaluators may take advantage of the Inverse Depletion Theory for Environmental Sampling Analysis (INDEPTH-ES) software developed by Oak Ridge National Laboratory and provided as an in-kind contribution to the IAEA by the USSP [12].

INDEPTH-ES accepts the measured uranium and plutonium isotopic composition of an irradiated material (e.g. data received from the NWAL) as an input and then performs a series of ORIGEN calculations to identify the irradiation scenario (i.e. parameters listed above) that best fits the measured data given a set of pre-selected reactor types. The resulting irradiation scenario can then be plotted with the measured data within the GUI for use within ES evaluation reports (see Figure 8).

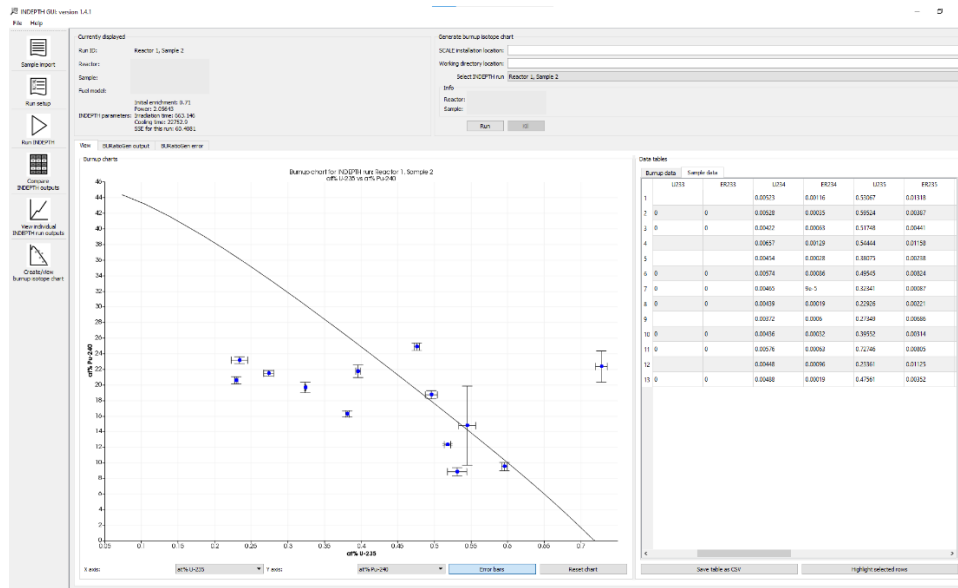


Figure 8. INDEPTH-ES GUI showing the identified irradiation scenario compared to measured data from an ES swipe sample

4.4 R-DAVE

Material characterization analysis of uranium samples (e.g. uranium ore concentrates) provides IAEA evaluators with information to verify States' declarations and investigate potential indicators of undeclared materials and activities. In particular, material characterization results (e.g. elemental impurity concentrations) can be used to assess material provenance to verify the completeness of States' declarations regarding source material [13]. To assist in the evaluation of such multidimensional impurity data, LLNL has recently provided the IAEA with another iteration of the stand-alone version of their Discriminate Analysis Verification Engine (DAVE). The current iteration (R-DAVE) is written in R and provides multiple machine learning algorithms that predict the most probable origin of a uranium material (of those within the reference dataset) or identifies it as a novel unknown (i.e. not originating from a class within the reference dataset); this provides IAEA evaluators with additional information to assess the declared origin of the sampled material. Additionally, the R Shiny GUI provides several data tables and interactive visuals that can be used within evaluation reports (see Figure 9). Collaboration is ongoing between LLNL and the IAEA to further refine R-DAVE considering the evaluator needs.



Figure 9. Examples of visualizations available within the R-DAVE GUI

5. Software developed by contractors

5.1 ESEE

The Environmental Sampling Environment Enhancement (ESEE) project is a multi-year effort to re-engineer the ESDB and develop a new ES evaluation platform. The current ESDB is based on obsolete technology, is both difficult and expensive to maintain, and does not support several types of analytical data received from the NWAL. As such, in order to make the evaluation process more efficient and to prepare for a potential increase in the number of samples collected in the field, the ESEE project will completely overhaul the ESDB and lead to the development of a new Web-based platform to be used by IAEA evaluators that will streamline all aspects of the ES evaluation process by, for example, providing a more user-friendly GUI, integrating NFC modelling and data analysis/visualization tools (e.g. those mentioned above), providing access to State declared information and nuclear material accountancy data, and incorporating a mechanism that facilitates the review, clearance, and distribution of reports.

6. Conclusions

The introduction of new software tools and data analysis techniques, as well as updates to existing software tools, has provided the IAEA with the necessary resources to leverage and investigate the multiple types of data received from the NWAL and to streamline the evaluation processes. These efforts continue to support the timely delivery of high-quality ES and material characterization analysis data evaluation reports to the Divisions of Operations, which are an important factor when drawing conclusions regarding the absence of undeclared nuclear materials and/or activities in a State. Additionally, the planned modernization of the ES evaluation platform is intended to further improve the efficiency of sample analysis evaluations.

7. References

1. IAEA Safeguards Glossary, 2001 Edition, International Nuclear Verification Series No. 3, Vienna 2001, section 9, pg. 72.
2. S. Richter, H. Kuehn, Y. Aregbe, M. Hedberg, J. Horta-Domenech, K. Mayer, E. Zuleger, S. Buerger, S. Boulyga, A. Koepf, J. Poths, K. Matthew, Improvements in routine uranium isotope ratio measurements using the modified total evaporation method for multi-collector thermal ionization mass spectrometry, *J. Anal. At. Spectrom.*, 2011, 26, 550-564.
3. C. Brennan, C. Szakal, D. Simons, J. Fassett, IAEA Safeguards Symposium, 2018, IAEA-CN-267/319.
4. C. Szakal, D. Simons, J. Fassett, A. Fahey, Advances in age-dating of individual uranium particles by large geometry secondary ion mass spectrometry, *Analyst*, 2019, 144, 4219-4232.
5. D. Fischer, R. Higgy, M. Nikkinen, Environmental sampling for detecting undeclared nuclear materials/activities, INMM/ESARDA Workshop, 2005.
6. M. Nikkinen, R. Räsänen, Cluster analysis for TIMS particle data analysis, IAEA Safeguards Symposium, 2006, IAEA-CN-148/118.
7. K. Vilece, C. Norman, J. Baute, G. Giaveri, M. Kiryu, M. Pellechi, Visualization of environmental sampling results at inspected facilities, INMM Annual Meeting, 2012.
8. K. Vilece, M. Schoeppner, A. Larson, M. Nikkinen, C. Norman, M. Lafitte, J. Baute, A new tool for the evaluation of environmental samples: development of software for morphological and elemental data evaluation, INMM/ESARDA Joint Annual Meeting, 2021.
9. M. Williams, S. Skutnik, I. Gauld, W. Wieselquist, R. Lefebvre, ORIGAMI: a code for computing assembly isotopics within ORIGEN, SCALE Code System 6.2.3, 2018, ORNL-TM-2005/39, 5.188–5.233.
10. C. Weber, I. Huff, J. Lefebvre, MSTAR 2019 code description and user's manual, 2020, ORNL/SPR-2019/1411.
11. US Support Programme to IAEA Safeguards. Task D2447: statistical methods for improved evaluation of environmental sample data.
12. B. Grogan, M. Purves, J. Lefebvre, INDEPTH for environmental sampling analysis (INDEPTH-ES): user guide (v1.4.1), 2018, ORNL-TM-2018/993.
13. M. Peńkin, S. Boulyga, D. Fischer, Application of uranium impurity data for material characterization in nuclear safeguards, *J Radioanal Nucl Chem*, 2016, 307, 1995-1999.