An Integrated Nuclear Archaeology Approach to Reconstructing Fissile Material Production Histories

Malte Göttsche[1], Baptiste Mouginot[2]

[1] Program on Science and Global Security, Princeton University, Princeton, NJ, USA
[2] Department of Engineering Physics, University of Wisconsin-Madison, Madison, WI, USA

Abstract:

Independent estimates assume that the world-wide civilian and military fissile material stocks amount to 500 tons of plutonium and 1,400 tons of highly-enriched uranium (HEU). To enable deep cuts in warhead arsenals, states will very likely need to verify declarations of fissile material stocks, as they could be used to build new nuclear weapons. One approach is nuclear archaeology, reconstructing the past production of those materials. Research has so far focused on specific measurements in reactors to assess past plutonium production, on deposits in gaseous diffusion plants and isotopics of depleted uranium tails to assess HEU production. For the large historical production of separated plutonium and HEU by Russia and the United States, uncertainties corresponding to large numbers of significant quantities remain with these techniques. A combination of measurements, fuel cycle simulations and reviews of records from past production activities could significantly reduce these uncertainties. This combination also would enable cross-checking information and measurement results for consistency. To demonstrate this approach, we conduct a case study. A declared production history is generated using the CYCLUS nuclear fuel cycle simulator. The simulation results are signatures resulting from the simulated history, some of which could in principle be measured. Assessing the isotopic compositions of waste streams, for example, can allow for cross-checking of declared fuel cycle histories. Based on this case study, the capabilities of the approach will be examined.

Keywords: nuclear archaeology; disarmament; fuel cycle simulations

1. Introduction

With the beginning of the Cold War, the United States and the Soviet Union launched their fissile material production (plutonium and highly enriched uranium, HEU) for military purposes. By the mid-1950s, both countries were already making ton-quantities of fissile material per year to supply their nuclear arsenals. They were soon joined by the United Kingdom (1951), France (1955), China (1964), and Israel (1965)—and later by India, Pakistan, and finally North Korea. According to independent estimates by the International Panel on Fissile Materials, there exist about 505 tons of plutonium and 1370 tons of HEU world-wide today [1].

Most large-scale fissile-material production programs were driven by a sense of urgency and typically shrouded in secrecy. It is generally believed that accounting for these military operations was poor. The fissile material production uncertainty is very large, and even states themselves have had difficulty reconciling production records with physical inventories. In the United States, for example,

estimated plutonium acquisitions exceeded the actual inventory by 2.4 tons, but it is not clear if this material ever existed [2].

These uncertainties will have to be understood and reduced as further progress toward nuclear disarmament is made. In particular, a solid understanding of fissile-material holdings is needed to achieve a meaningful degree of predictability and irreversibility of future arms-control initiatives. Speculations about unaccounted fissile-material stockpiles, possibly equivalent to hundreds of nuclear weapons, could make progress in this area very difficult. To this end, states must be able to verify the past fissile material production [3].

2. An integrated nuclear archaeology approach

In order to understand and reduce the uncertainties in the amount of produced fissile materials, new methods and tools must be developed that help reconstruct the past fissile material production history and enable verification. This is called nuclear archaeology, a concept introduced already in 1990 [4]. The state of nuclear archaeology research is at a low level despite its importance. Some initial research has been conducted on measurement concepts for nuclear archaeology in uranium enrichment plants [5]. The isotopic composition of the depleted uranium tails can be assessed, to determine whether HEU had been produced [6, 7]. Other techniques under development include quantitative estimates of the plutonium production in graphite-moderated reactors (GIRM) [6, 8, 9] and heavy water reactors [10] by examining the graphite moderator or structural reactor elements. By examining isotopic ratios of trace elements, the neutron fluence can be determined, which yields the total amount of plutonium produced in the reactor, assuming the reactor design to be fully known. Some further unpublished and perhaps classified research appears to have been conducted by U.S. government scientists [11]. Of all published research, only GIRM has been experimentally validated to a larger extent.

All past research results only deal with examining particular fuel cycle facilities in isolation. However, the nuclear archaeology toolbox could be much broader. Additional signatures could be sought. What is lacking is a systematic and integrated approach that ties together all available information – not only from measurements, but also from available records about the past fissile material production. Such an approach could be used to identify inconsistencies (for example between records and today's measurements), help understand the underlying reasons for the current uncertainties, and reduce them.

Some nuclear weapon states have a large number of nuclear facilities involved in the fissile material production. The operations of these facilities changed over time (e.g. changes in the power levels of the reactors), as did the complex material transfers between these facilities. For example, spent fuel has sometimes been reprocessed and used as feed in enrichment plants. Enrichment operations also had a level of complexity. For example, natural uranium has sometimes been enriched to low enriched uranium in one plant, which has subsequently been fed into another plant to produce HEU [12, p. 57].

To calculate the material flows in such complex fuel cycles, an integrated fuel cycle simulation tool would be very useful. Data on facility operations provided to the inspector or plausible assumptions could be used as input to simulate the nuclear materials as they pass through the fuel cycle. The simulation results of such forward-modelling would tell about the various signatures to expect from what is available today, for example the isotopic composition of different types of wastes (e.g. depleted uranium tails or radioactive reprocessing waste). It could then be checked whether information gathered today, such as waste measurements, are in agreement with the simulation results. In particular in more complex fuel cycles, various signatures are correlated, and examining

these correlations itself can provide information on the production history, e.g. to which extent reprocessed uranium was used for HEU production.

The joint evaluation of measurements and simulations based on provided data can potentially reduce the uncertainties of the fissile material estimates beyond what is possible based on either approach alone. Some documented data may be inaccurate, or different recorded data maybe inconsistent in itself, for example because of past inventory measurements with large uncertainties. Measurements today can perhaps help resolve such issues.

In the following, we first examine a tool that could be developed for the forward-modelling of complex fissile material production histories. Second, we use a case study to demonstrate the integrated nuclear archaeology approach.

3. Forward-modelling simulation tool

The forward-modelling simulation tool must be able to handle all relevant types of nuclear facilities. It must be able to calculate the composition and masses of nuclear materials at the different fuel cycle stages. To manage the complex fuel cycle histories, the code must provide for options to change facility operations over time, introduce new facilities and shut down old ones at specified times, and change the material flows between facilities. To realize this, discrete fissile material flows must be calculated, as opposed to continuous material flows resulting from a fuel cycle in a steady state.

The Cyclus fuel cycle tool [13] is an open source, agent based fuel cycle simulator. Fundamentally, the Cyclus simulator tracks the discrete flow of materials between facilities over time. It can also provide time series data such as material inventories, incorporating radioactive decay. Facilities are represented as agents. Each agent has its own unique and independent behaviour. The agents do not communicate directly with one another, but interact through the Dynamical Resource Exchange (DRE), which calculates the fissile material transfers between them.

At the beginning of each time step, each facility sends its own material requests to the DRE. The DRE then collects the corresponding bids made by the different facilities that have material to offer. Finally, the DRE solves the market problem by finding a solution that matches requests with bids, allowing the materials to flow between the different facilities. Each facility can specify requested quantities or compositions of materials, and they can decline trade if the offered materials do not meet its own requirements (e.g. regarding isotopic composition).

The Cyclus suite is offered as a pair of libraries: the Cyclus core contains the DRE as well as input and output interfacing, while the Cycamore library provides a basic set of nuclear facility agents. The separation between the simulation agents and the solver core provides flexibility and customizability. The Cyclus development team supports the development of the Cycamore plugins, which correspond to low fidelity archetypes that incorporate some physics [14]. If the Cycamore archetype suite does not capture a desired behavior, contributors can design their own specialized facilities as plugins.

For example, a modified version of the Cycamore enrichment facility has been developed for this research [15]. The original Cycamore enrichment facility allows enriching the uranium-235 content of a feed stream. All minor isotopes (those apart from uranium-235 and uranium-238, for example uranium-234 or uranium-236) are directly sent to the tails. This does not represent the physical reality, where also the minor isotopes are being enriched. To enable the tracking of additional isotopes, the new enrichment archetype allows for manually specifying the enrichment of isotopes other than uranium-235 and uranium-238 as well, and calculates the correct isotopic composition of the product and tails

streams. To enable this, the ratios of each isotope's product to feed enrichments must be specified manually.

Given a desired product mass $M_{product}$ and uranium-235 enrichment τ_p, as well as the specified uranium-235 content in the tails τ_t, the enrichment agent will first compute the required feed mass M_{feed} , which also depends on the uranium-235 enrichment of the feed τ_f, using

$$M_{feed} = M_{product} * \frac{\tau_p - \tau_t}{\tau_f - \tau_t}$$

The tails mass results from mass conservation. The concentrations of the specified minor isotopes in the product are then calculated based on their content in the feed and the specified enrichment ratios. The remaining minor isotope masses from the feed that are not in the product are transferred to the tails.

3. Demonstration of the integrated approach

In the following, we use a case study to provide an example of an integrated nuclear archaeology assessment, using more than one measurable signature, documentation provided to the inspector and forward-modelling to learn about the past fissile material production. For the purpose of clarity, we keep the case study simple. While complex fuel cycle computer simulations are not absolutely required in this simple case, a realistic nuclear weapons state case could be much more complex, in which case a complex simulation tool would be required to forward-model the fuel cycle based on the provided documentation.

For the case study (see Fig. 1), we assume a state used natural uranium to produce plutonium in reactors and a reprocessing plant (Fig. 1, path A) and HEU in a gaseous diffusion enrichment plant (path B). Because the state assumed limited access to natural uranium and wanted to maximize use of its natural uranium resources, it decided to enrich some of the reprocessed uranium (which still contains 0.6% to 0.7% uranium-235) to produce HEU (path C).

The principal goal is to determine how much HEU and plutonium the state produced, based on three pieces of information that were declared: the total amount of natural uranium used (208 tons), information on the reactors (full reactor designs known, specific power 4.45 W/gIHM and fuel burnup 300 MWd/t), and knowing the HEU enrichment (94%). Notably, to quantify the plutonium and HEU production, the material transfers corresponding to the paths A, B and C must be quantified.

While simplified, this case is relevant. For example, the United States released data on its history of natural uranium purchases, and further data on its nuclear program. [16]. This data has already been used to conduct a rough consistency test [17]. Also, at least the Soviet Union and the United States re-used their reprocessed uranium in reactors to produce more plutonium or in enrichment plants for the production of HEU [12, p. 57].

In the following, we propose an approach to solve this nuclear archaeology case study in three parts, combining measurements with forward-modelling.

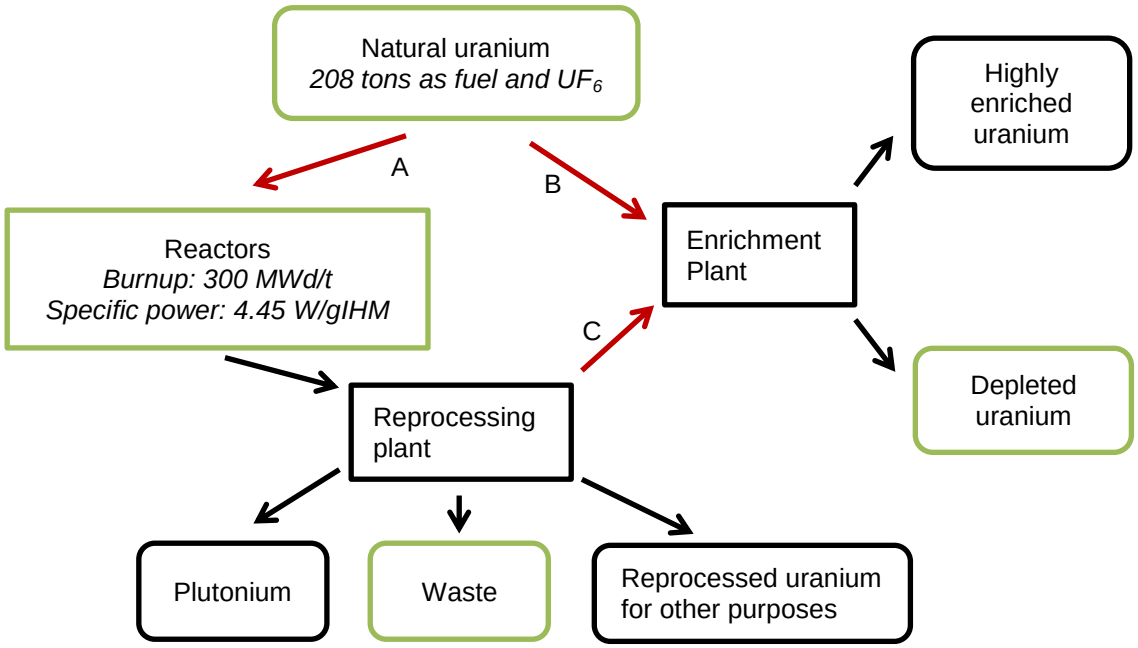

Figure 1: The fuel cycle of the case study. Certain knowledge from declared information or measurements exists for the materials and facilities marked in green. The italic text refers to information declared to the inspectors. The case study focuses on how to use the available data to gain information on the plutonium and HEU production, by quantifying the material transfers indicated by the red arrows, marked A, B and C.

i) The amount of produced plutonium and the required natural uranium (path A) can be determined by taking samples from structural components of the reactor cores for forensic analysis of trace elements.

ii) Confidence in the declared information can be increased by performing consistency-checks. As an example relating to the plutonium production part, the declared fuel burnup can be checked by forensic analysis of the radioactive (high-level) reprocessing waste (for example liquid waste using inductively coupled plasma mass spectrometry). Some plutonium remains in the waste – in particular from military reprocessing activities several decades ago [18]. The ratio of plutonium-239 to plutonium-240 indicates the fuel burnup, if the reactor design is known (see for example graphs in [19, p. 159]). It would be obtained by depletion calculations (forward-modelling). Because of the long half-lives, this signature is very little dependent on the time passed since the fuel discharge from the reactor, which may be unknown.

iii) To distinguish between HEU production from natural uranium feed (path B) and reprocessed uranium feed (path C), forensic analysis by thermal ionization mass spectrometry of samples taken from a number of depleted uranium tails containers could be used. Uranium-236 is produced from uranium-235 when irradiated in reactors, by capturing a neutron [12, p. 58]. Therefore, both HEU as well as the corresponding depleted uranium tails contain uranium-236, if the uranium had previously been irradiated. Given a reprocessed uranium feed, the concentration of uranium-236 in the depleted uranium tails depends on the uranium-235 enrichment of the produced HEU and the tails. It can be computed by using the matched abundance ratio or M* (M-star) cascade theory [20, 21, 22].

Natural uranium, in contrast, contains no uranium-236. Hence, the extent to which natural and reprocessed uranium were used can be determined by measuring the uranium-236 content.

5. Demonstration of the integrated approach: Results

i) Knowing the reactor design and the fuel burnup, infinite lattice depletion calculations using MCODE [23] were performed. The lattice used here corresponds to a reactor design similar to the Savannah River Site (USA) reactor, containing natural uranium slugs and lithium-aluminum alloy control rods. The MCODE simulations yield the isotopic composition of the spent fuel. The uranium isotopics of the spent fuel are shown in Table 1. The MCODE simulations also show that the reactors produced 0.25 g of plutonium per kg of uranium fuel.[1] In the future, a capability to perform such calculations could be integrated into Cyclus. For now, we have manually included the MCODE results in our Cyclus simulation.

Isotope	wt-%
Uranium-234	$5.30 \cdot 10^{-3}$
Uranium-235	0.678
Uranium-236	$5.13 \cdot 10^{-3}$
Uranium-238	99.312

Table 1: Uranium isotopics of the spent fuel

For the purpose of this paper, we assume (without actually assessing the ratios of trace elements in the reactors' structural elements) that it was successfully found that the reactors produced a total of 42 kg of plutonium, using 168 tons of natural uranium (path A). Given that the inspectors know that overall 208 tons of natural uranium were used for both plutonium and HEU production, they now know that 40 tons of natural (un-irradiated) uranium were used for HEU production (path B).

ii) The MCODE simulations show that, at 300 MWd/t, the plutonium isotopic ratio is $Pu-239/Pu-240 = 62.81$. If the forensic analysis of radioactive waste yielded this ratio, the measurement results would independently confirm the declared information that was used for forward-modelling.

iii) For a uranium-235 product enrichment of 94 wt-%, a tails enrichment of 0.3 wt-%, and the uranium-236 feed content from Table 1, we obtain a uranium-236 tails content of $3.85 \cdot 10^{-3}$ wt-% according to the M* cascade theory.[2] These values are used in the Cyclus simulation.

If the uranium-235 tails enrichment is unknown, it would be obtained from the forensic analysis of the tails. In this case study, it is 0.3 wt-%. Let us assume that the average uranium-236 content of the tails the inspectors measured is $2.14 \cdot 10^{-3}$ wt-%. At these values, the relative measurement uncertainties using Thermal Ionization Mass Spectrometry can be smaller than 10^{-3} [24], so the measurements have sufficient precision at these low concentrations. It can be deduced that about 44% of the enrichment feed was natural uranium, and 56% were reprocessed uranium. Thus, 50 tons of reprocessed uranium were enriched (path C). Note that it is necessary to measure the tails, *and* to know the uranium-236 content of the feed, which was obtained from forward-modelling based on the burnup data. Results would have been wrong, if the declared burnup had been wrong. By using forensic analysis of reprocessing waste, however, this cheating scenario could be excluded.

As the main purpose of the case study is only to demonstrate the nuclear archaeology concept in general, we have not conducted a comprehensive uncertainty analysis. The results are therefore preliminary.

[1] The results of the MCODE simulations are presented in order to demonstrate the proposed nuclear archaeology concept. We have validated the reactor model used for MCODE only to a limited extent.
[2] We have not compared the calculated uranium-236 concentrations against experimental values. Therefore, the computed concentration is used in this paper only to explain the described nuclear archaeology concept.

6. Conclusions

Using a case study, this paper has shown, how assessing several indicators of past fissile material production (here: isotopics of depleted uranium tails, isotopics of the reprocessing waste, and documentation provided to inspectors) can be used to study the consistency of data and deduce additional information that had not been provided. It has also shown the importance of combining computer simulations and measurements.

Cyclus provides a good framework for a fuel cycle simulation tool, as it calculates discrete fissile material flows and allows for changes in the fuel cycle design and facility operations over time. For example, a more complicated version of the case study would be a fuel cycle history with a much larger number of facilities with different operational characteristics each, changing over time.

The current facilities that exist as part of the Cycamore library have only very limited capabilities. To comprehensively use Cyclus in the nuclear archaeology context, such libraries must be significantly extended, for example allowing to use a wider range of operational parameters that might be part of provided documentation as simulation input.

Once the integrated nuclear fuel cycle code for nuclear archaeology has been developed, it must be validated, preferably against historical data. The code structure of Cyclus is modular, which facilitates validation: Facility agents can be programmed and implemented without changing other facility agents or the kernel. Therefore, specific extensions or new agents may not require repeated validations of the overall code, if the other parts have been previously validated.

With regard to the integrated nuclear archaeology approach, a robust uncertainty analysis of the gained information on the past fissile material production history will be central. For example, measurement uncertainties must be included. Uncertainties are also introduced by sampling strategies, e.g. from how many depleted uranium containers samples are taken, as isotopic ratios may vary between them. Information on past activities will be incomplete, which introduces uncertainties of the fuel cycle simulation results due to uncertainties of the input parameters. A study should be conducted comprehensively studying the various uncertainties and their propagation.

Overall, this approach can be able to identify errors (for example in the provided documentation) that would not have been found without consistency-checks, thereby possibly increasing precision of fissile material estimates. Also, it may enhance the capability to detect deliberate attempts to declare false or incomplete production histories. Such a widened nuclear archaeology approach could significantly contribute to enable a solid understanding of fissile-material holdings, thereby increasing predictability and irreversibility of future arms-control initiatives.

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References

[1] International Panel of Fissile Materials, “Global Fissile Material Report 2015,” 2015.

[2] U.S. Department of Energy, “The United States Plutonium Balance, 1944-2009,” 2012.

[3] A. Glaser and M. Göttsche, “Fissile Material Stockpile Declarations and Cooperative Nuclear Archaeology,” in *FM(C)T Meeting Series, Verifiable Declarations of Fissile Material Stocks: Challenges and Solutions*, United Nations Institute for Disarmament Research Resources, 2017.

[4] F. v. Hippel, “Warhead and Fissile-material Declarations,” in *Reversing the Arms Race: How to Achieve and Verify Deep Reductions in the Nuclear Arsenals*, New York, Gordon and Breach Science Publishers, 1990.

[5] S. Philippe and A. Glaser, “Nuclear Archaeology for Gaseous Diffusion Enrichment Plants,” *Science & Global Security,* pp. 27-49, 2014.

[6] S. Fetter, “Nuclear Archaeology: Verifying Declarations of Fissile-Material Production,” *Science & Global Security 3,* pp. 237-259, 1993.

[7] M. Sharp, “Applications and Limitations of Nuclear Archaeology in Uranium Enrichment Plants,” *Science & Global Security 21,* pp. 70-92, 2013.

[8] C. Gesh, “A Graphite Isotope Ratio Method Primer - A Method for Estimating Plutonium Production in Graphite Moderated Reactors,” Richland, Washington, USA, 2004.

[9] B. Reid, D. Gerlach, P. Heasler and J. Livingston, “Trawsfynydd Plutonium Estimate,” Richland, Washington, USA, 2009.

[10] A. Gasner and A. Glaser, “Nuclear Archaeology for Heavy-Water-Moderated Plutonium Production Reactors,” *Science & Global Security 19,* pp. 223-233, 2011.

[11] T. Wood, B. Reid, C. Toomey, K. Krishnaswami, K. Burns, L. Casazza and L. Duckworth, “The Future of Nuclear Archaeology: Reducing Legacy Risks of Weapons Usable Material,” *Science & Global Security,* pp. 4-26, 2014.

[12] International Panel on Fissile Materials, “Global Fissile Material Report 2009: A Path to Nuclear Disarmament,” 2009.

[13] K. Huff, M. Gidden, R. Carlsen, R. Flanangan, M. McGarry, A. Opotowsky, E. Schneider, A. Scopatz and P. Wilson, “Fundamental concepts in the Cyclus nuclear fuel cycle simulation framework,” *Advances in Engineering Software 94,* 2016.

[14] R. W. Carlsen, M. J. Gidden, M. B. McGarry, A. C. Opotowsky, A. Scopatz and P. Wilson., ““Cycamore v1.3.0.” Figshare,” http://dx.doi.org/10.6084/m9.figshare.1427430.v1, 2015.

[15] B. Mouginot, ““CYCHT v0.1.0.” Figshare,” https://doi.org/10.6084/m9.figshare.4909334.v1, 2017.

[16] T. B. Cochran, W. M. Arkin, R. S. Norris and M. M. Hoenig, Nuclear Weapons Databook Vol.2, U.S. Nuclear Warhead Production, Cambridge, MA, USA: Ballinger, 1987.

[17] F. v. Hippel, “Consistency Tests for the Declarations of U.S. Fissile-Material Production,” *Science & Global Security 19,* pp. 1-14, 2011.

[18] R. Alvarez, “Plutonium wastes from the U.S. nuclear weapons complex,” *Science & Global Security 19,* 2011.

[19] International Panel on Fissile Materials, “Global Fissile Material Report 2010: Balancing the Books. Production and Stocks,” 2010.

[20] A. de la Garza, G. Garrett and J. Murphy, “Multicomponent Isotope Separation in Cascades,” *Chemical Engineering Science 15,* 1961.

[21] A. de la Garza, “A Generalization of the Matched Abundance-Ratio Cascade for Multicomponent Isotope Separation,” *Chemical Engineering Science 18,* 1963.

[22] H. G. Wood, “Effects of Separation Processes on Minor Isotopes in Enrichment Cascades,” *Science & Global Security 16,* 2008.

[23] Z. Xu, “Design strategies for optimizing high burnup fuel in pressurized water reactors,” Dissertation, Massachusetts Institute of Technology, Cambridge, MA, USA, 2003.

[24] S. Richter, H. Kühn, J. Truyens, M. Kraiem and Y. Aregbe, “Uranium hexafluoride (UF6) gas source mass spectrometry for certification of reference materials and nuclear safeguard measurements at IRMM,” *Journal of Analytical Atomic Spectrometry 28,* pp. 536-548, 2013.