Uncertainty Assessment of the Isotope Ratio Method in Nuclear Archaeology

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Abstract

In the context of nuclear disarmament, robust verification mechanisms to assess the completeness of fissile material baseline declarations are lacking. In the field of nuclear archaeology, the Isotope Ratio Method (IRM) has been proposed as a way to use measurements of materials in the long-lasting structures of shut-down reactors to infer the quantities of produced plutonium. We propose a method of assessing the systematic uncertainties associated with IRM, focusing on incomplete information relating to the reactor operation as well as uncertainties in nuclear cross-section data. We develop a model to reconstruct local plutonium production from a measurement of Ti-48/Ti-49 given a set of input parameters regarding the assumed operation. The application of this reconstruction model is demonstrated for a CANDU 6 pressurized heavy-water reactor model. With two different estimates for input parameter uncertainties, we use quasi-Monte Carlo methods to estimate the output uncertainty of the model. The single value estimates of the local plutonium production in the considered scenarios have an uncertainty of 4 to 11 %, which illustrates that initial uncertainty assessments can have a significant impact on the output uncertainty. Statistical tolerance intervals are proposed to interpret the uncertainty in a manner that is meaningful to verification purposes.

1 Introduction

Nuclear armed states' assessments of their own fissile material production histories include significant uncertainties [1]. In the United States as of 2009, for example, production records indicated that 2.4 tons of plutonium were produced in addition to what was known to be in the inventory [2]. Decreasing these uncertainties would cause immediate safety and security benefits for the state itself and would, furthermore, be an essential condition for nuclear disarmament of any nuclear armed state: baseline declarations of weapons-usable fissile material inventories and the corresponding production histories will likely need to be issued and independently verified [3].

The Isotope Ratio Method (IRM) can be used to estimate the lifetime plutonium production of shut-down reactors, and is therefore both a tool to reduce uncertainties of past production estimates, and to verify fissile material baseline declarations [4]. Should inspectors return to North Korea, for instance, there would be interest in applying this method to the graphite-moderated reactor in Yongbyon, which produces the North Korean plutonium [5], [6].

Specifically, IRM is based on assessing the neutron fluence $\Phi = \int \phi dt$ (where ϕ is the neutron flux) using isotopic measurements of trace impurities in several samples that underwent neutron activation, taken in various locations within or very close to the core of shut-down reactors. This method has been developed for graphite-moderated reactors, where samples from the graphite would be taken (GIRM) [7], and has been proposed for heavy water reactors [8]. Elements with relevant isotopes include boron, lithium, chlorine, calcium, titanium, chromium, iron, nickel, zirconium, and lead [8].

A detailed understanding of the uncertainties of the plutonium estimates resulting from the IRM is crucial, to assess whether IRM measurements are consistent with what a state knows about the reactor's plutonium production, or what it declares to inspectors. A detailed quantitative uncertainty study for GIRM has been conducted by Heasler et al. [9]. For a generic scenario, they find an expected relative uncertainty of 1.62 %. The paper finds that by far the largest contribution to the overall uncertainty stems from incomplete knowledge of parameters related to the reactor design and its operational history[1].

The analysis in [9] assumes that best-estimate values of the design and operational parameters can be obtained. It then takes some uncertainty around this best estimate into account. We argue, however, that the knowledge in particular on the operational history may in some cases be somewhat limited, such that a reliable estimate of the operational parameters is hard or impossible to obtain.

We present a quasi-Monte Carlo based approach for assessing IRM uncertainties on a case-by-case basis. Using extensive simulations we propagate several different sources of uncertainty through the IRM algorithm to quantify the uncertainty of a plutonium estimate.

2 Uncertainties of the Isotope Ratio Method

Deducing plutonium production of a reactor requires three steps. First, the neutron fluence at a specific location in the reactor must be expressed as a function of the measured isotopic ratio of samples at that location. According to the depletion equation, this depends on the production and depletion paths of the considered isotopes. It also depends on the corresponding one-group reaction cross-sections, which depend on the neutron energy spectrum at that location. This can be obtained from neutron transport simulations. Second, the plutonium production at a specific location must be expressed as a function of the local fluence estimate. For this, fuel burnup calculations are required,

[1] In Heasler et al., this design and operational uncertainty is referred to as the "Reactor Physics Error".

which require input parameters such as reactor power or discharge burnup. Third, the global plutonium production of a reactor must be inferred from local plutonium production. This requires full-core reactor simulations to obtain spatial information.

With all of these three steps, various kinds of uncertainties and errors are introduced, which all must be propagated through the calculation process to obtain the final uncertainty on the plutonium estimate. In [10], possible sources of the IRM uncertainties have been identified. They include nuclear data, design and operational parameters of the reactor, model approximations and computational errors. In [9], the major contributions to the uncertainty on design and operational history are in descending order: fuel pin radius, fuel temperature, graphite density, equivalent boron concentration, graphite temperature and specific power.

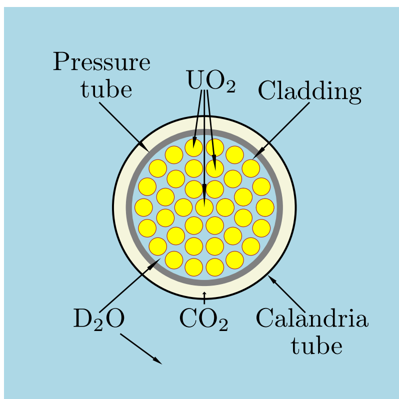
In this study, we address only the first two steps of the IRM. To perform the sampling-based uncertainty quantification, we construct a computational model to estimate the local plutonium production from a hypothetical isotopic ratio measurement. This model is explained in the following section. The uncertainty sources are implemented as input parameters of the model that can be varied initially, but remain constant during a single execution.

To illustrate the method, we focus on two categories of input uncertainties: operational parameters and nuclear cross-section data. We assume the reactor and fuel design to be known, for instance from visual inspections. Regarding the operational history, which we assume to be known to various degrees, we focus on the influence of uncertainties in fuel burnup, reactor power and moderator temperature. Nuclear cross-section data uncertainties are considered for the intuitively most relevant nuclides. U-238 and Pu-239 are the prevalent isotopes in the reactor fuel whose nuclear reactions account for the plutonium production and removal rate. Therefore, we consider the uncertainties of the induced fission (n,fis) and radiative neutron capture (n,γ) cross-sections of both isotopes. When considering IRM specifically, the cross-sections of the indicator elements are important. We demonstrate our method using Ti-48/Ti-49 as indicator, since the ratio has been used in other studies and is considered to be suitable for conducting nuclear archaeology on reactors with a high fluence. Thus, the reaction cross-sections relevant to the development of these ratios are also included in our parametrization of the input uncertainties. In general, suitable isotopic ratios need to be identified carefully [11], before conducting a comprehensive uncertainty analysis.

In the next section we explain our computational model, which takes a value of Ti-48/Ti-49 as well as a set of input parameters and estimates the local plutonium production for a given reactor model.

3 Local Plutonium Reconstruction Method

As explained previously, we focus on the first two steps of the IRM, i.e., local fluence and plutonium estimates. Fig. 2 illustrates the logical steps of the reconstruction model. We use the validated continuous-energy Monte-Carlo reactor physics code `SERPENT 2` [12] to simulate neutron transport and calculate fuel depletion. The neutron transport simulation yields the neutron energy spectrum ϕ_E and the fuel depletion calculation computes the plutonium production. Since we limit ourselves to local plutonium reconstruction, we perform infinite lattice simulations to represent a single fuel channel in the center of the reactor. Fig. 1 shows the geometry of the generic CANDU 6 fuel channel, which we implemented in `SERPENT 2`.



Lattice pitch[a]	28.6 cm
Pressure tube ⌀	10.338 cm
Fuel element ⌀	1.206 cm
Cladding thickness	0.92 cm
Elements per bundle	37
Fuel composition	nat UO_2
Cladding material	Zircaloy 4[b]
Coolant	D_2O
Moderator material	D_2O

[a] The lattice pitch is the distance between fuel channels
[b] Zr–1.5%SN–0.2%Fe–0.1%Cr

Figure 1: Fuel channel of a generic CANDU 6 reactor. The permanent structures interesting for nuclear archaeology are the pressure tube and the calandria tube.

The input parameter *burnup* determines the duration of one simulation, which in our model acts as one batch of fuel being burned and subsequently extracted to obtain plutonium. We refer to this as one production campaign. During a simulation, the neutron energy spectrum $\phi_{E,i}$ is tallied for each burnup step at the relevant location in the reactor geometry, i.e., where we expect to measure an isotopic ratio. To derive neutron spectrum related quantities, i.e., neutron flux and neutron fluence, we use the time-averaged neutron spectrum $\overline{\phi_E}$, which is computed by averaging over all burnup steps of a simulation. Integrating over energy yields the average neutron flux $\overline{\phi}$ and integrating over time yields the neutron fluence Φ_0 of a single batch. The produced plutonium is obtained as a mass density by summing over the individual mass densities of each plutonium isotope in the depleted fuel, plus isotopes of other elements which decay into plutonium with an extremely short half-life, e.g., U-239 and Np-239.

Our implementation of step one of the IRM, the evolution of the isotopic ratio, i.e., Ti-48/Ti-49 , is expressed explicitly as a function fluence and respective one-group cross-sections. The isotopic ratio is derived from the isotopic vector of the indicator elements, which is given as:

$$\vec{N}(t) = \exp(\boldsymbol{A}t)\vec{N}(0). \tag{1}$$

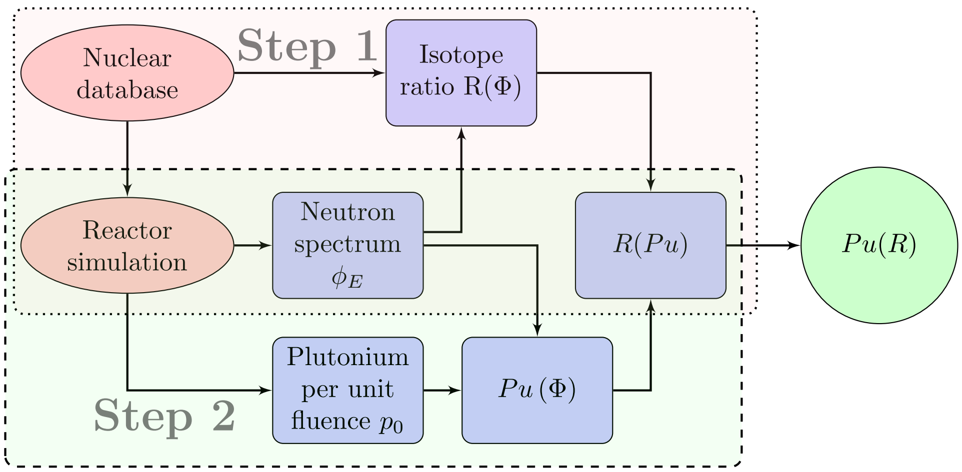


Figure 2: The logical steps of the reconstruction method for local plutonium production.

Here $\boldsymbol{A}$ is the transition matrix containing the respective one-group cross-sections and the average neutron flux. We calculate one-group cross-sections

$$\Sigma_i = \frac{\int \sigma_i \phi\,(E)\,dE}{\int \phi\,(E)\,dE}$$

for the relevant nuclear reactions using energy-dependent cross-section data. We use ENDF/B-VIII.0 [13] cross-section data obtained from the JANIS 4.0 database [14]. In Fig. 2 we highlight the dependence on fluence, as this quantity will create the link to plutonium production.

Step two creates the relation between local fluence and plutonium production. We define the plutonium per unit fluence for a single batch as:

$$p_0 = \frac{Pu}{\Phi_0}.$$

To estimate long-term plutonium production, i.e. plutonium produced by multiple subsequent production campaigns, we assume that the reactor is operated under near identical conditions for each batch. This produces the following simplifications for the framework: the average neutron flux is constant the entire time and the plutonium per unit fluence is constant. Thus, the long-term plutonium production is calculated as:

$$Pu\,(t) = p_0\,\overline{\phi}\,t, \tag{2}$$

since $\overline{\phi}$ is constant, $\Phi = \overline{\phi}\,t$ and the above expression is equal to $Pu(\Phi)$ in Fig. 2.

Combining Equations 1 and 2, the isotopic vector is given as:

$$\vec{N}(Pu) = \exp(\boldsymbol{A}\frac{Pu}{p_0\overline{\phi}})\vec{N}(0). \tag{3}$$

Finally, we obtain the desired isotopic ratios by dividing the respective components of the vector and then seek the inverse, i.e. the plutonium production corresponding to a specific isotopic ratio. We use the numerical tools in Python to invert the relationship.

Thus, the computational framework describes the relationship between long-term plutonium production in one fuel channel and an isotopic ratio measurement, based on a computational reactor model and a set of operational parameters. Given an isotopic ratio value, the model returns an estimate for the produced plutonium.

4 Uncertainty Quantification

To propagate uncertainty sources through the model we use quasi Monte Carlo methods. That is, we parametrize the model in terms of uncertainty sources and use quasi random sampling to select input parameter values from the multidimensional parameter space. The boundaries of the input parameter space reflect the assessment of initial uncertainty. In Table 1 we present the two different input parameter spaces we use in this analysis for a generic CANDU 6 reactor. In addition to the range, the underlying distribution is determined, which later affects the sampling algorithm. In the cases we present, the nuclear cross-section uncertainties follow a normal distribution and the operational parameter uncertainties are uniformly distributed. The former is due to the nature of the measurement and evaluation process of nuclear cross-section libraries, the latter reflects our assessment that lack of information may require an equal consideration of all possible values. With our selection of examples, we showcase how the uncertainty assessment of each parameter can differ between cases and that the impact on output uncertainty can be significant.

To create samples, we use the Sobol sequence generation algorithm explained in [15] with the initialization numbers from [16]. This is a low-discrepancy sequence, the advantage of which is that it covers a multidimensional parameter space evenly with a lower number of samples compared to pseudo-random sampling algorithms. The Sobol sequence itself only generates uniformly distributed values. To obtain samples of the normally distributed parameters mentioned above, we first generate uniform samples for the entire parameter space with the Sobol sequence and then transform the pertinent dimensions into a normal distribution using the inverse cumulative distribution function of the normal distribution.

To apply the reconstruction model, an isotope ratio value is required. Since real measurement data is not available, we determine a range of reasonable values for Ti-48/Ti-49 , using separate simulations, and perform the reconstruction for several values along that range. This estimation is based on an approximation of the total fluence in a CANDU reactor with a lifetime power production of the Bruce 1 reactor in Canada.

To assess the output uncertainty of the model, 4000 sets of input parameters are sampled and the model is evaluated for each one, i.e. for one isotope ratio

Table 1: Estimated input parameter uncertainties

	CANDU-1 (C-1)	CANDU-2 (C-2)
Temperature[a]/ K	343–353	333–363
Thermal power / MW_t	2000–2500	1000–2500
Burnup / $MW\,d\,kg^{-1}$	0.2–1.5	0.2–4.0
σ $(n,\gamma)^{47}Ti$ [b]	±3%[c]	±6%[d]
σ $(n,\gamma)^{48}Ti$	±3%	±6%
σ $(n,\gamma)^{49}Ti$	±3%	±6%
σ (n,fission) ^{238}U	±1.2%	±2.4%
σ (n,γ) ^{238}U	±1.3%	±2.6%
σ (n,fission) ^{239}Pu	±1.4%	±2.8%
σ (n,γ) ^{239}Pu	±4.3%	±8.6%

[a] Only the temperature of the moderator liquid (D_2O) is varied.
[b] σ refers to the *one-group cross section.*
[c] Uncertainties are given as an interval of the one relative standard deviation around the expected value.
[d] For CANDU-2 we doubled the relative standard deviation to showcase the impact of larger overall uncertainties.

value, the reconstruction framework computes 4000 different plutonium values. This process is repeated for each case. The basic characteristics of the resulting plutonium distribution, such as the sample mean and the sample standard deviation, are easily calculated numerically. To characterize the distribution in manner that is meaningful for nuclear archaeology, we propose *statistical tolerance intervals*. The tolerance interval contains a certain proportion p of the population with a confidence level γ and give upper and lower bounds on the expected plutonium production, rather than a best estimate value. As the distribution of plutonium values is not easily parametrized, we choose a non-parametric approach using order statistics to calculate the intervals. Such an approach is explained in [17]. The results are presented in the following section.

5 Results

In Fig. 3 we compare the two different cases of the CANDU reactor model. The left graph in the figure shows that the assessment of input parameter uncertainty affects the width of the tolerance interval: higher input uncertainty leads to higher output uncertainty. It also shows that the mean value of the distribution is affected by range of input parameters. The former reflects the precision of the reconstruction; the latter pertains to its accuracy. While we do not make a quantitative assessment of the accuracy, it nevertheless shows that both aspects are influenced by the choice of input parameters. The right graph in the Fig. 3 indicates a negligible increase of relative uncertainty. The relative uncertainty here is quantified as half the width of the tolerance interval relative to the mean of the interval. For a 95% confidence interval, this relative half-width is approximately double the relative standard deviation.

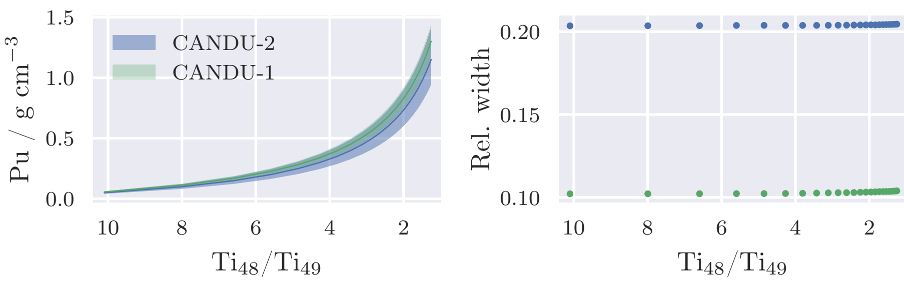


Figure 3: Reconstructed plutonium with the CANDU reactor model. The shaded area indicates the 95% tolerance interval. The right graph displays half the width of the interval relative to the interval mean. The range of titanium ratios corresponds approximately to the expected lifetime of a CANDU power reactor.

Since we perform infinite lattice simulations, the plutonium is quantified in units of $\mathrm{g\,cm^{-3}}$. A rough estimate of the total plutonium amount can be made by multiplying with the volume of the reactor fuel elements. This does not take the inhomogeneity of the neutron flux into account and should only be used as a rough approximation. Results scaled to the reactor volume are shown explicitly for a selected ratio value in Tab. 2. The table shows a best estimate with standard deviation and two different tolerance intervals. The mean value and the standard deviation are useful quantities to describe any distribution, however, the uncertainty quantified by the standard deviation is often evaluated under the assumption that the quantity to be measured follows a normal distribution. In Table 2, the parametric tolerance interval assumes the plutonium values are normally distributed and is derived from the sample mean and standard deviation. The non-parametric tolerance interval gives different upper and lower bounds on the plutonium estimate. Since we assumed some uniformly distributed input parameters, a non-normally distributed output is to be expected, if the respective input parameters have a high impact on the output. This effect highlights why a more thorough assessment of tolerance limits, beyond those generally implied by a mean value and a standard deviation, may be warranted, especially when the verification of declarations and (non-)compliance with a treaty are at stake.

Table 2: Estimate for plutonium from hypothetical ratio measurements. The 95% tolerance interval have a 95% confidence level. For illustration purposes the plutonium densities are scaled by the entire reactor fuel volume.

Ratio	Case	$\overline{Pu} \pm \sigma$	Parametric[a]	Non-parametric
10.1	C-1	$548 \pm 29(5\%)$kg	490–607 kg	495–607 kg
	C-2	$484 \pm 55(11\%)$kg	374–595 kg	397–595 kg
4.2	C-1	$3292 \pm 177(5\%)$kg	2939–3645 kg	2971–3645 kg
	C-2	$2909 \pm 332(11\%)$kg	2246–3572 kg	2386–3572 kg
1.3	C-1	$12620 \pm 686(5\%)$kg	11250–13990 kg	11370–14000 kg
	C-2	$11154 \pm 1278(11\%)$kg	8603–13700 kg	9140–13700 kg

[a] Assuming normally distributed Pu values.

6 Conclusion

In this paper we have demonstrated a Monte-Carlo based approach to uncertainty propagation for the Isotope Ratio Method. While we have used a specific set of parameters to showcase the method, more parameters can be added to the computational model to take other sources of uncertainty into account. Furthermore, we propose statistical tolerance intervals as a useful measure to quantify the uncertainty, as they give a more robust statement about the expected plutonium production. The different test cases demonstrate that the uncertainty of a local plutonium production estimate is strongly dependent on the initial assessment of input uncertainty. Going forward, we are using variance based sensitivity analysis to study the relative impact of different input parameters on the output uncertainty.

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