An Integrated Approach to Nuclear Archaeology

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Abstract:

As of today, mainly two approaches to reconstruct past fissile material production can be considered: First, some concepts for measurements in shut-down facilities and of radioactive wastes have been suggested that could be taken today to gain information about the past. However, there are cases where the production history cannot be reconstructed with low uncertainty based on measurements alone. Second, information from documentation of past nuclear facility operations can be used as input to simulation tools to quantify the produced fissile materials. However, information from such records will have uncertainties; there might even be missing information. Here, we propose to expand this toolbox with an approach that combines the different methods for an integrated assessment. Taking data from various sources into account directly allows for consistency-checking. Furthermore, using Bayesian inference and numerical methods, more reliable estimates of parameters relating to the fuel cycle history can potentially be obtained - at least for small nuclear programmes.

Keywords: Disarmament, Forensics, Verification, Bayesian Inference, Nuclear Archaeology

1. Introduction

While there is extensive experience in verifying both the correctness and completeness of nuclear material declarations issued by non-weapon states that are members of the Non-Proliferation Treaty, there is a lack of methods to verify nuclear material “baseline” declarations, i.e. the first verified declaration a state makes upon entering an agreement. 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 [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].

In addition to direct data on produced fissile materials, such records would contain operational information of the nuclear facilities. This paper will focus on plutonium, even though highly enriched uranium is a similar challenge. The methods proposed here can likely also play a role for the uranium context. For reactors, in addition to reactor and fuel designs, operational information would include data on reactor power, fuel burnup, and cooling time (which refer to the time passed since a specific campaign). We call these data operational parameters.

In order to obtain more accurate fissile material inventory estimates, a first approach in reconstructing the production history is performing reactor simulations with more accurate codes than those used decades ago. One such code is SERPENT 2 [3]. Such codes would take the operational parameters as input, and produce the isotopic composition of the discharged fuel – including plutonium, but also fission products and minor actinides – as output.

In addition, measurements in shut-down facilities and of radioactive waste can be taken to obtain complementary data. This approach is known as nuclear archaeology.

What is lacking however, 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), reconstruct missing data from records using measurements, and quantify and reduce the uncertainties on the amount of produced fissile materials.

The challenge is two-fold. First, the measurement toolbox should be expanded to allow for assessing various complementary signatures. For plutonium, in addition to looking at reactor samples, we propose to measure concentrations of high-level reprocessing waste and assessing the total high-level waste mass, for instance using calorimetry or antineutrino measurements. Second, a data analysis approach is required to examine all available data from records and measurements to maximize the capability of consistency-checking, information reconstruction and uncertainty reduction. Here, we propose to use Bayesian inference for this purpose.

Section 2 provides an overview of the state of research and discusses new measurements for nuclear archaeology. Section 3 discusses the proposed Bayesian approach, and shows results from a proof-of-concept study related to combining high-level waste measurements and data from records into a joint assessment. Section 4 presents first thoughts for the extension of this method to a fully integrated assessment taking the proposed measurement techniques into account.

2. The Measurement Toolbox

The state of nuclear archaeology research with regard to plutonium is at a low level despite its importance. Most developed is a technique in which samples from shutdown reactor cores are taken and, through analysis of their isotopic ratios of trace elements, an estimate of the neutron fluence can be obtained, from which the plutonium production can be deduced. One example of this approach is the Graphite Isotope Ratio Method, which is applicable for graphite-moderated reactors [4,5,6]. While this method has been tested, similar methods for heavy water reactors are in an early state of development. A concept has been proposed which can be used in reactors that use aluminum tubes, such as the Indian CIRUS reactor [7]. However, if such facilities have already been decommissioned when inspection activities are agreed upon, these techniques will not function. Another challenge are facilities that have been used for various purposes, e.g. the production of both plutonium and tritium. A method has been proposed that measures two isotopic ratios in order to deduce both the total fluence and the contribution of plutonium production to that fluence [8].

High-level reprocessing waste could be used as a complementary technique to obtain an estimate of the total produced mass of plutonium, as the plutonium mass correlates with the volume/mass or heat emission of the radionuclides in the total waste. We propose to study antineutrino measurements [9] and calorimetric methods in this context. If several methods can be carried out, they could be used to reduce uncertainty or to check for consistency. Inconsistencies may point at additional hidden waste, or that the reactor structure that samples have been taken from have not been in the reactor over its full lifetime. It may also be that not all measurements are possible, for instance that reactors have already been dismantled before verification occurs.

Third, measurements of isotopic concentrations of high-level reprocessing waste could be used [10]. It contains nearly all fission products and minor actinides after dissolving the spent fuel. Accordingly, it contains a rich isotopic signature of past fuel cycle activities. This waste could be used to compare results with the operational history contained in records, reconstructing parts for which records are incomplete or contain uncertainties. This would improve the accuracy of deducing plutonium production from reactor simulations. Furthermore, the measurements could serve to check a declaration containing the operational history for consistency. If a state declared that a reactor was used for civilian purposes with high burnup, this method could prove that low burnup campaigns for possibly military purposes were run. Similarly, a reactor may have run for more time than declared, which could be detected by examining the cooling times. While it is not clear whether the proposed approach can be applied to complex programmes, it may be feasible for use for simpler programmes of a complexity similar to the North Korean case.

3. Bayesian Inference for Nuclear Archaeology

To be able to conclude how much plutonium (or also highly enriched uranium) has been produced, and whether an according declaration is consistent and plausible, a data analysis approach that is able to tie together information from various sources including records must be pursued. Bayesian inference is well-suited for this purpose, as it allows both integrating measurements and information from records into one integrated assessment.

To demonstrate this approach, this section focuses on determining operational parameters by combining information from records and measuring isotopic concentrations of reprocessing waste. It presents a first and preliminary proof-of-concept study using very simple scenarios.

3.1. Theoretical Background

Our task is to solve an inverse problem. The isotopic composition of high-level waste ($\vec{y}_{obs}$), which would be measured, is the output of reactor simulations. We seek, however, the input to those simulations – which we call forward-simulations –, this being the operational parameters ($\vec{x}$). The forward-simulations can then be thought of like a model that can compute $\vec{y} = f(\vec{x})$. Reactor simulations couple a Monte Carlo neutron transport routine with a fuel depletion routine. Therefore, $f(\vec{x})$ cannot be described by a simple function, which could perhaps be inverted analytically. Instead, we invert it by Bayesian inference using numerical methods.

Bayesian inference can solve the inverse problem by treating it statistically. It is in particular suited for inverting intractable and complex models, as is the case here. It calculates the *posterior*, which is the distribution of the probabilities $p(\vec{x}|\vec{y}_{obs})$ that specific reactor parameter combinations $\vec{x}$ could have produced to measured isotopic composition $\vec{y}_{obs}$, using Bayes' theorem

$$p(\vec{x}|\vec{y}_{obs}) \propto p(\vec{y}_{obs}|\vec{x}) \cdot p(\vec{x})$$

$p(\vec{y}_{obs}|\vec{x})$is the *likelihood*, which is the distribution of probabilities that the measured isotopic composition would have been obtained by a specific combination of operational parameters. The output of a forward-simulation $\vec{y}$ is compared to the measured isotopic composition $\vec{y}_{obs}$. We assume that $\vec{y}$ is normally distributed, hence

$$p(\vec{y}_{obs}|\vec{x}) = \prod_{i=1}^{N} exp\left(\frac{-|y_{obs,i} - f_i(\vec{x})|}{\sqrt{2}\sigma_i}\right)^2$$

where the index i represents an isotope under consideration, and σ_i is the corresponding uncertainty, which must be chosen. It must include all sources of uncertainties: measurement uncertainties, model uncertainties, etc. The equation holds if the isotopes chosen are independent of each other under the measurement technique.

The particular benefit of this approach is that *prior* knowledge can be included, which is given by manually formulating $p(\vec{x})$. This could, for instance, be information from records of the production history. It does not have to be taken at face value, as an uncertainty can be associated with the information. According the Bayes' theorem, this prior information is then combined with the measurement to produce the posterior. Another advantage of this approach is that, due to its probabilistic nature, it allows for the propagation of uncertainties, so that we obtain the uncertainty of the reactor parameter estimates.

To numerically estimate the posterior, we explore the reactor parameter space by evaluating different reactor parameter combinations $\vec{x}$. For this, we use Markov Chain Monte Carlo [11]. Due to a large number of required simulations, using a high-fidelity model – such as a detailed reactor simulation like SERPENT 2 – can be computationally prohibitive. Therefore, we have developed a surrogate model that accurately represents the high-fidelity model but is computationally feasible to evaluate. Specifically, we run 1000 SERPENT 2 simulations with different parameter combinations $\vec{x}$ and interpolate using Gaussian Process Regression [12,13]. Details of the approach are discussed in [14].

3.2. Implementation

The reactor design we use in this study is an infinite lattice model of the Savannah River Reactor K's inner core with the Mark 15 fuel design (1.1% enrichment) using data from [15], [16], [17], [18], [19], see also [14]. This reactor was designed for the production of weapons-grade plutonium. We limit our proof-of-concept study to reconstructing hypothetical values of fuel burnup (B) and cooling time (Ct).

For this purpose, we use ^{137}Cs, ^{154}Eu, ^{95}Mo, ^{142}Nd and ^{90}Sr. We choose these for their good sensitivity on the two parameters, which has been quantified by a variance-based sensitivity analysis, see [14]. In an actual application, one would study ratios of isotopes. If the proof-of-concept can be demonstrated with individual isotopic concentrations, it should also be feasible with isotopic ratios, as the underlying mathematical principles would remain the same.

We consider a very limited plutonium production scenario with a state having a single reactor, discharging and reprocessing its fuel only once or twice. The high-level waste is stored in a single tank. Conceptually, this very roughly corresponds to the situation in North Korea, when IAEA inspectors conducted on-site-inspections after the state joining the Non-Proliferation Treaty, including reprocessing waste samples [20].

To solve the inverse problem, first, $\vec{y}_{obs}$ must be calculated. We do this by choosing specific values of $\vec{x}$ we use as input to SERPENT 2 to produce $\vec{y}_{obs}$. We then use the software package PyMC3 [21] to calculate the posterior. The algorithm has no knowledge of the chosen values, but reconstructs them based on $\vec{y}_{obs}$. For each case study, we have used 40,000 posterior evaluations.

Three case studies have been designed (see also Table 1 for the priors of these cases and Table 2 for the corresponding values of the parameters to be inferred):

1. This study seeks to demonstrate that simultaneously taking prior information (e.g. from records) and measurements into account by Bayesian inference results in reduced uncertainties of estimates compared to examining both data sources in isolation. We show this by comparing two scenarios.
   <u>Scenario 1</u>: A single fuel batch has been reprocessed, the burnup and cooling time are unknown. This is implemented using broad uniform probability distributions of the two parameters as a prior, i.e. giving equal probability for the parameters within a large range defined by minimum and maximum values, $U[min, max]$. A vector y_{obs} for the isotopes ^{137}Cs, ^{154}Eu, ^{95}Mo, ^{142}Nd and ^{90}Sr is calculated. We assume that all isotopic concentrations carry an uncertainty of 5% (σ_i).
   <u>Scenario 2</u> is the same as the first case, but now assuming prior information e.g. from authenticated records, resulting in narrower ranges of the uniform prior.

2. This study seeks to demonstrate the Bayesian inference method for a case where some information is contained in records, but it is incomplete. We attempt to reconstruct the missing information. We again compare two scenarios:
   <u>Scenario 1</u> corresponds to scenario 1 of case study 1, but now there is also a second batch with different burnup and cooling time values. The batches ($\vec{y}_{obs,j}$) result in waste of composition $\vec{y}_{mix}$ being stored in the same tank, $\vec{y}_{mix} = \alpha\vec{y}_{obs,a} + (1-\alpha)\vec{y}_{obs,b}$. We assume no prior information on the mass ratio α of the two batches, and on the values of the second batch.
   <u>Scenario 2</u>: The two batches are the same as in scenario 1. While there is still no prior information on the second batch or α, we now implement a more informed prior for the first batch.

3. This study seeks to demonstrate the Bayesian method in identifying inconsistencies, pointing to false information in a declaration or records. The scenario is the same as the previous scenario, but there is now a prior on the second batch. It has a uniform high probability in a false region (corresponding to false information from records).

Scenario	Burnup (MWd/Kg)	Cooling Time (Years)	Mass Ratio α
Case study 1 Scenario 1 Scenario 2	$U[0,3]$ $U[1.80,1.85]$	$U[0,50]$ $U[28.5,30.5]$	
Case study 2 Scenario 1 Scenario 2	a: $U[0,3]$, b: $U[0,3]$ a: $U[1.7,1.9]$, b: $U[0,3]$	a: $U[0,50]$, b: $U[0,50]$ a: $U[9,11]$, b: $U[0,50]$	$U[0.1,0.9]$
Case study 3	a: $U[1.7,1.9]$, b: $U[2,3]$	a: $U[9,11]$, b: $U[0,1]$	$U[0.1,0.9]$

Table 1: Comparison of priors for the three case studies. For case studies 2 and 3, each scenario includes priors for two batches and a mass ratio α.

Scenario	Simulated Measurement
Case study 1 Scenario 1 Scenario 2	Burnup = 1.835 MWd/Kg Cooling Time = 29.43y
Case study 2 Scenario 1 Scenario 2 **Case study 3**	Burnup a = 1.835 MWd/Kg Burnup b = 0.8 MWd/Kg Cooling Time a = 10.4 y Cooling Time b = 30.2.y Mixing Ratio = 0.4

Table 2: Parameter values used for the inference process

3.3. Results & Discussion

3.3.1. Case study 1: Uncertainty Reduction, Scenario 1

We reconstruct the burnup and cooling time for a single batch of reprocessing waste based on simulations of isotopic measurements. Figure 1, illustrates the results obtained.

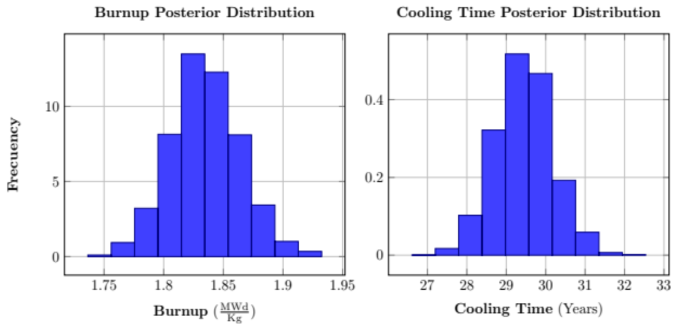

Figure 1: Case Study 1, Scenario 1 marginal posterior distributions for burnup and cooling time

As can be observed from Figure 1, both burnup and cooling time have been successfully reconstructed, with relative uncertainties of about 1.5% and 2.5%, respectively.

3.3.1. Case study 1: Uncertainty Reduction, Scenario 2

In this scenario we look now on the effects of a constrained prior on the inference process. This simulates the presence of additional information (e.g. operational records) in the reconstruction. Figure 2, shows the results obtained

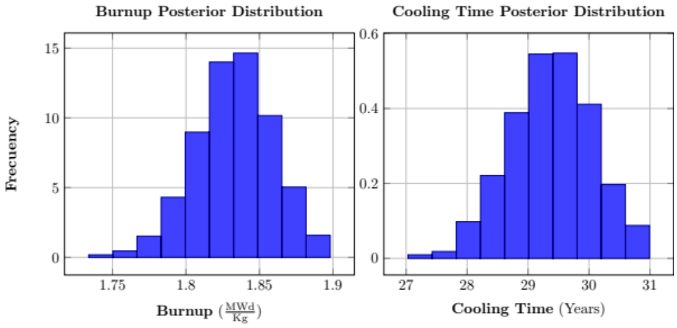

Figure 2: Case Study 1, Scenario 2 marginal posterior distributions for burnup and cooling time

This time the spread of the posterior distributions is now smaller than in Scenario 1 with a relative uncertainty for burnup and cooling time of 1.2% and 1.9% respectively. This means that through the inclusion of additional information, the uncertainty on the parameter prediction can be reduced.

3.3.2. Case study 2: Reconstructing Missing Information, Scenario 1

In this scenario we aim to reconstruct the burnup, cooling time and the mixing ratio of two batches of reprocessing waste using priors based on the known limits of the parameter values. Figure 3 illustrates the results obtained.

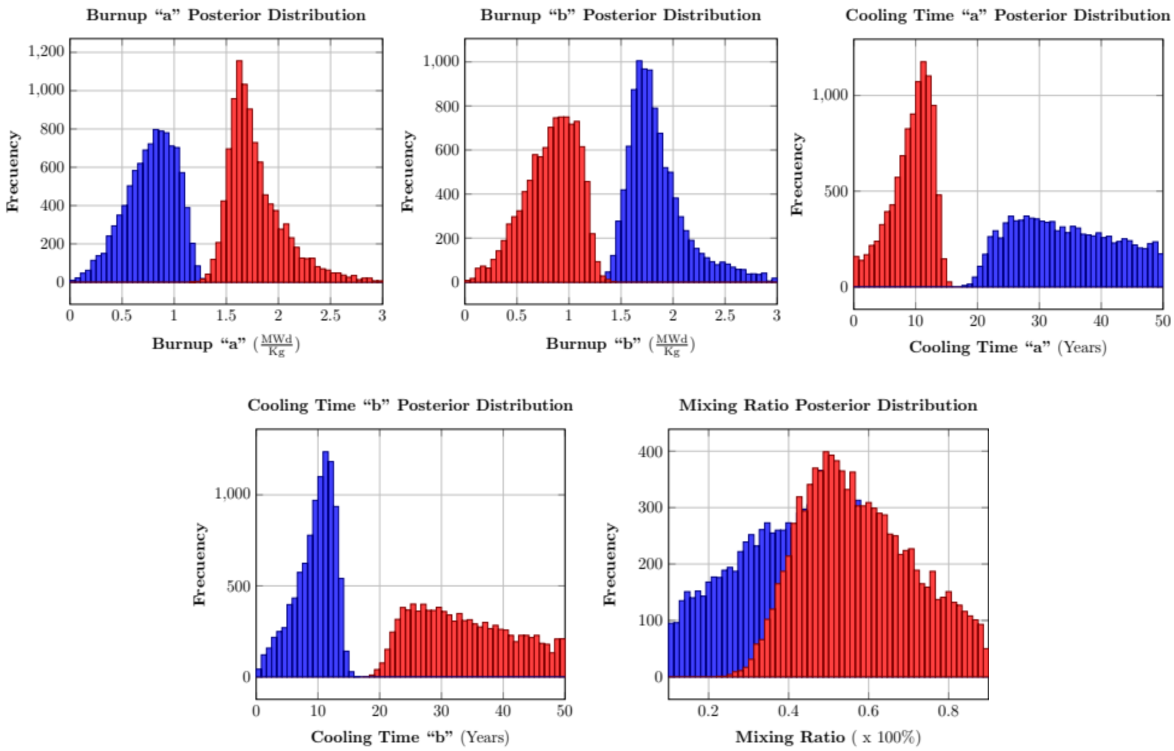

Figure 3: Case study 2, Scenario 1 posterior distributions. Two solutions are found (blue and red).

It can be observed that now two sets of possible solutions have been determined marked in blue and red. It can be seen that the two solutions are almost symmetric (a small asymmetry being due to numerical reasons), and both are correct: In both cases, the two batches are correctly characterized, the only difference lies in the attribution of batch "a" and "b". The relative uncertainties for the reconstruction of the parameters burnup a, burnup b, cooling time a, cooling time b and the mixing ratio for the red solution are 15.9%, 29.5%, 35.5%, 23.6% and 35% respectively. In comparison to case study 1, we now observe significantly larger reconstruction uncertainties.

3.3.2. Case study 2: Reconstructing Missing Information, Scenario 2

Similar to scenario 1, in this case, however, prior information on batch 1 is available and is included in the problem in the way of a constrained prior. Figure 4 shows the results obtained.

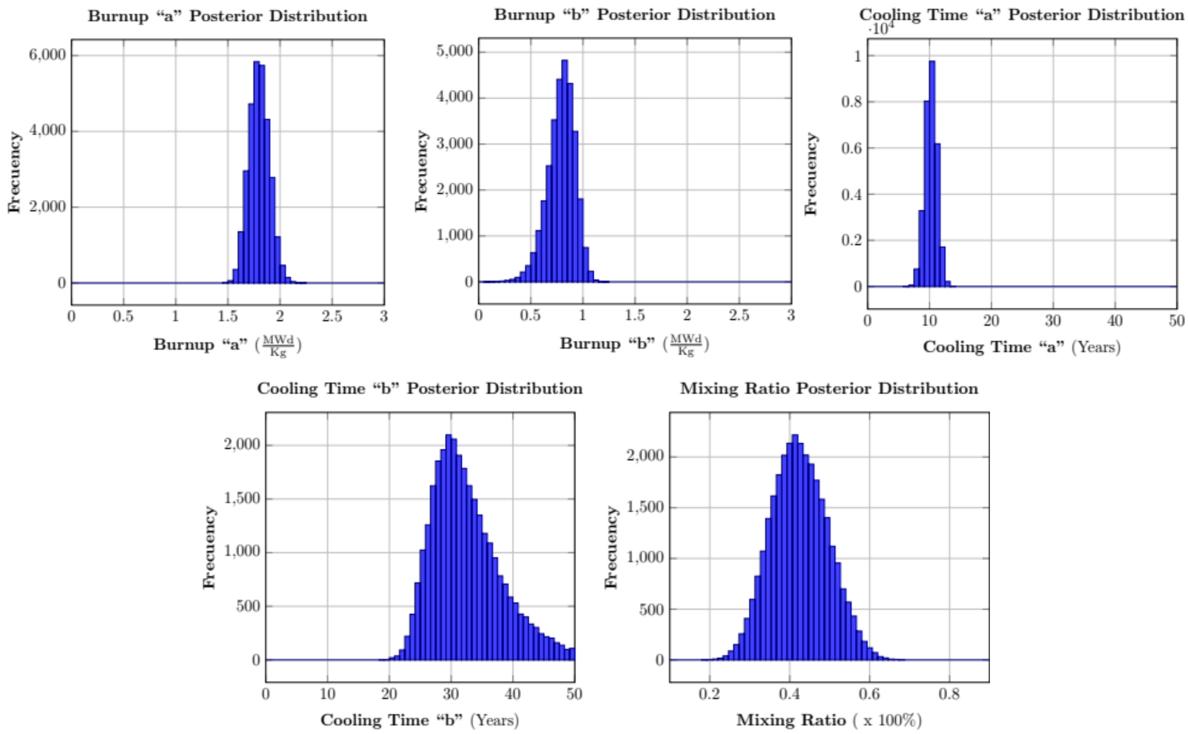

Figure 4: Case study 2, Scenario 1 posterior distributions. The previous multimodal solution has become unimodal, with the means of the distributions corresponding to the actual values of the target solution

It can be now seen that in comparison to scenario 1, only one solution has been found for each of the parameters. The relative uncertainties corresponding to the parameters burnup a, burnup b, cooling time a, cooling time b and mixing ratio are 5.4%, 16.7%, 9.4%, 16.9% and 16.6% respectively. It can then be concluded that through the use of extra information (e.g. operational records etc.), codified through priors, uncertainty on the reconstruction of all parameters can be reduced. This includes specifically also the batch where no additional information is provided. Information on batch a dramatically reduces the uncertainties for batch b. One can also see that the true data is very close to the maximum likelihood result for all parameters.

3.3.3. Case study 3: Consistency-Checking

In this scenario we simulate the effects of false information provided by a declaring state for the case of a mixture of two batches. This has been included in the priors for the second batch. Figure 5 shows the output of the inferred variables:

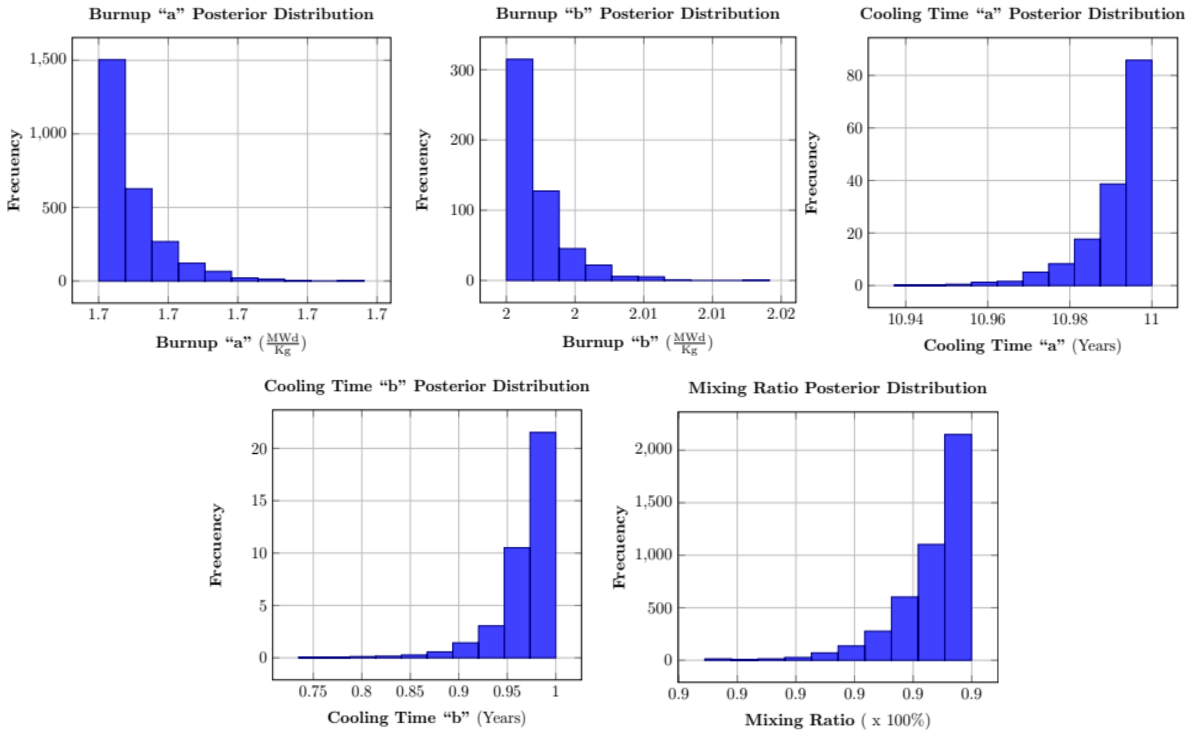

Figure 5: Case study 3, Posterior Distributions. The shape of the distributions points towards the expected solution as being outside of the parameter range described by the priors used

As it can be observed in Figure 5, the posterior distributions point towards the typical set being outside of the space defined by the priors, thus making apparent the inconsistency and irregularity brought upon by the false information present in the priors or by proxy, declarations.

4. Integrated Assessment

The case studies demonstrate, that Bayesian inference can in principle be used to infer parameters of interest and to quantify their uncertainties. However, all considered scenarios have a very limited complexity, i.e. a low number of parameters to be reconstructed. Future research on the waste context should focus on extending our approach to handle more complex scenarios, and thereby study its capabilities and limitations. Here, we have used only few isotopes. In a more involved application, a much larger number of isotopic ratios could be used, which could in principle allow for deducing additional parameters.

In particular, the number of reprocessed batches, each with varying operational parameters, would be larger. For small nuclear programmes such as North Korea, perhaps this larger list of parameters could still be reconstructed as a much larger set of isotopes could be considered in the analysis than those presented here. However, this would clearly not work for large programmes such as in the U.S. or Russia. In such cases, strategies must be found to reduce the amount of parameters to be reconstructed. This is called model reduction, a classical topic in computational science.

While there are methods of model reduction where the resulting reduced number of parameters that describe a physical system are themselves non-physical -as is the case for principal components analysis-, in this application they must remain physical. For instance, if inconsistencies related to specific parameters are identified, only physical parameters would allow for a clarification. One way to do this is through the use of copula priors. Copulas are joint probability distributions of two or more variables which encode the likelihood of specific combinations of these variables. For example in a one reactor system, if the operation with simultaneous high power and low temperature was not possible, a negligible probability can be assigned a priori to such combinations, thus reducing the complexity and number of possible solutions. A similar procedure could be applied in the case of two or more batches, given for example the knowledge -through historical records or expertise- that these could not have been produced between certain years, an appropriate copula can be constructed.

A second research topic is to examine to which extent this approach could also be useful in the context of different nuclear archaeology measurement techniques. It appears applicable in examining samples from reactors. While the original approach measures one isotopic ratio to reconstruct one parameter (neutron fluence), an implementation has already been proposed using two isotopic ratios for two parameters, as explained above. This method may be further expanded in complexity, which would motivate a Bayesian assessment in this context.

While the various possible measurements can be used as stand-alone assessments, lastly, it should be researched how they –together with the documentation- could be integrated into a joint assessment. Two examples: First, there exist uncertainties in translating measured neutron fluence in a reactor into a plutonium estimate. One such uncertainty source may be missing knowledge on discharge burnup. Relevant information could be found from high-level waste measurements, which could then be taken into account. Second, different measurement techniques can reconstruct the same parameter. For example, both measuring neutron fluence and measuring antineutrino rates to deduce the total amount of produced waste inform about plutonium production.

In the quest for such integrated approaches, statistical methods play a role. The combination of predictions from different realizations or models (waste isotopic measurements, antineutrino measurements, etc.) of a given output such as burnup, can be done in its simplest form through a weighted sum of the predictions of each model. A generalization of this can be found in the field of Data and Sensor Fusion through the use of a so called steady-state Ensemble Kalman Filter [22]. In this approach, the combined estimation for the parameter $\bar{x}$ can be expressed as a function of each model's likelihood $\pi_Y(Y_{obs}|x_i)$ and prediction x_i, where i denotes various measurements, for M models in the following way :

$$\bar{x} = \sum_{i}^{M} w_i x_i$$

Where the weights w_i are calculated:

$$w_i = \frac{\pi_Y(Y_{obs}|x_i)}{\sum_{j=1}^{M} \pi_Y(Y_{obs}|x_j)}$$

These weights have to be estimated through comparison between the different model outputs and the real observations. They can be interpreted as the degree of belief -and such of uncertainty- on the estimate provided by its corresponding analysis method.

In addition to this, the variance of the combined estimate can computed:

$$\sigma^2 = \sum_{i=1}^{M} w_i (x_i - \bar{x})^2$$

Provided new and better estimations of x_i are available, this filter can be applied recursively, thus resulting in an updated combined prediction for such parameters.

Acknowledgements

We thank Florian Wellmann (RWTH) for his expertise in Bayesian inference. This work has been funded by the Volkswagen Foundation.

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