VERIFYING INITIAL DECLARATIONS USING NUCLEAR ARCHAEOLOGY METHODS

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Abstract

When concluding Comprehensive Safeguards Agreements, states submit initial declarations. To reach the Broader Conclusion under Additional Protocols, the IAEA evaluates all available information for consistency which can include access to facilities and historical operating records. For states that had extensive fuel cycles prior to initial inspections, this is a complex undertaking, as the IAEA visits to South Africa have shown for example. One important instance where such activities would need to be pursued in the future is if IAEA inspectors were to return to the DPRK. In this paper, we propose the application of nuclear archaeology methods in such contexts, which could expand the IAEA's technical capabilities by allowing for a systematic examination of the history and the identification of inconsistencies in a timely manner. Specifically, various forensic measurements can be used to check for coherence with provided records. Taking the DPRK's plutonium program as an example, we present on-going technical research to first discuss how measuring isotopic ratios in high-level reprocessing waste – where available – can be used to probe declarations using statistical approaches. Second, we show how information on historical reactor operations can be deduced by measuring trace elements from samples (e.g. graphite) taken inside reactor cores, discussing the existing GIRM technique and proposing a novel statistical analysis which can enhance capabilities.

1. INTRODUCTION

When concluding a Comprehensive Safeguards Agreement (CSA), states must submit an initial declaration, which the IAEA then verifies for correctness and completeness. But in practice, only when Additional Protocols are also in place is the IAEA able to conclude that *all* nuclear materials within a state are being used for peaceful purposes. To reach this "broader conclusion," the IAEA evaluates all available information it has about a state, which includes information and access to facilities that the examined state must provide. Facility designs, research activities, information from inspections, and any other available information may contribute to this.

To issue a broader conclusion, the IAEA must have addressed all identified anomalies, discrepancies, and inconsistencies [1]. For states that had extensive fuel cycles before joining the NPT, confirming the correctness and completeness of initial declarations is a complex undertaking. In the 1990s, for example, the list of states with prior nuclear programs that signed a CSA included Argentina, Brazil, Kazakhstan, Lithuania, North Korea, South Africa, and Ukraine [2].

According to the IAEA, "Transparency in nuclear activities in a State increases the IAEA's understanding of the nuclear programme, facilitates analysis of its coherence and consistency, and ultimately increases confidence in the conclusions drawn by the IAEA for that State." [3] In principle, the IAEA can also demand historical operating records, and assess the internal consistency of the declared past, present and planned nuclear program [2]. It is not clear, however, that the IAEA systematically demands or analyses information about a state's past nuclear activities. The IAEA acquires confidence over time as safeguards are being applied in a routine manner, if no suspicions arise.

A complementary method is indeed to examine the history of fissile materials. By studying records and other sources of knowledge detailing past production and removals, one may calculate the inventories expected today, which can be compared to the fissile materials currently accounted for. Forensic measurements at shut-down fuel cycle facilities and of the stored waste would be an essential element. The field of reconstructing fissile-material history, known as nuclear archaeology, has been a topic of research since around 1990[4]. Indeed, a more systematic examination of the history could identify inconsistencies in a more timely manner.

There is one IAEA precedent for such an approach: when South Africa joined the NPT. In 1992 and 1993, South Africans granted the IAEA insight into thousands of pages of documentation, inter alia, about their past production of fissile material [5]. This allowed the IAEA to assess the size of the inventory of highly enriched uranium (HEU) that South Africa would be expected to possess, which can be obtained by re-calculating the production history.

One important instance where the IAEA might need to pursue such activities in the future is if inspectors were to return to the Democratic People's Republic of Korea. As has been noted in previous studies [6, 7], verified baseline declarations of fissile material holdings would be an essential element towards denuclearization. Therefore, this paper highlights the application of established as well as novel nuclear archaeology methods in this context.

2. DPRK'S NUCLEAR PROGRAM

North Korea's nuclear weapons program includes at least one centrifuge-based uranium enrichment plant for HEU production and a graphite-moderated nuclear reactor for plutonium production. Information about these facilities and their operating histories is roughly known from past IAEA inspections, North Korean declarations, and remote surveillance. Several publications have used this information to estimate the fissile material inventory, albeit without any means of corroboration. [8–10]

The remainder of this paper focuses on methods to determine the plutonium production and operating history of nuclear reactors, specifically the 5MWe reactor at Yongbyon, North Korea. Figure 1 illustrates the geometric layout of the 5MWe reactor core. The reactor uses natural uranium as fuel, CO2 gas as coolant, and graphite as moderator. It contains approximately 800 fuel channels, and each channel can hold up to 10 fuel elements. To estimate the plutonium production of this reactor, researchers use reactor-physics simulation software that simulates the neutron transport and the fuel depletion.

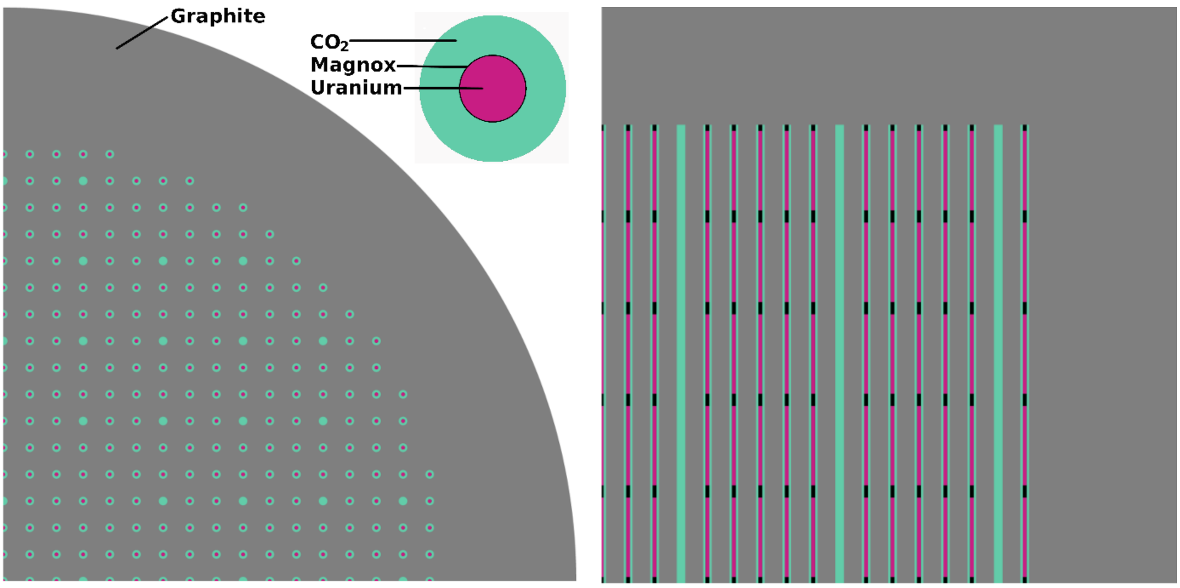

Figure 1: The image on the left shows a horizontal cross-section of a quarter of the 5MWe reactor core. Its top right corner shows a close up of a single fuel channel, with the CO_2 coolant, the Magnox cladding and the uranium fuel. The image on the right depicts a vertical cross-section of top half of the same section of the core. The reactor was implemented with geometry parameters given in Park and Hong [9].

Such software is a cornerstone of the nuclear archaeology methods described later in this paper, but first, we used it to construct a hypothetical case study of the 5MWe reactor with a known synthetic truth and simulated isotopic evidence. Using the reactor physics codes ONIX [11] and OpenMC [12], we simulated the irradiation of two separate batches of fuel in the reactor (see Table 1). In this scenario, the first batch of fuel is irradiated for 1095 days with an average thermal power of 8.4 MW. Over a period of 100 days, the entire core is discharged and sent to reprocessing, and fresh fuel is loaded into the reactor. This second batch of fuel is irradiated for 1785 days with an average thermal power of 16.7 MW, after which it is discharged and reprocessed. A year after the discharge of the second batch, inspectors are permitted to investigate the facilities and collect samples from the graphite in the reactor and the high-level waste storage tank but are prevented from accessing the reprocessed plutonium and uranium inventories.

This scenario imitates the situation in North Korea in 1994 and 1995, although in reality the discharged fuel was not reprocessed until much later [13]. It imagines that under the Agreed Framework, inspectors were allowed to collect samples to verify North Korea's past nuclear activities.

Table 1: Synthetic truth of the simulated operation of the 5MWe reactor. This scenario corresponds to the “pre-1994 discharge” scenario described by de Trouilloud de Lanversin and Kütt[10].

	Irradiation Time	**Cooling Time**	**Average Thermal Power**	**Average Burnup**	**Plutonium**
Batch A	1095 days	2250 days	8.4 MW	185 MWd/t	8.25 kg
Batch B	1785 days	365 days	16.7 MW	600 MWd/t	24.39 kg

3. TRADITIONAL NUCLEAR ARCHAOLOGY (GIRM)

We apply the Graphite Isotope Ratio Method (GIRM) to our ground truth, similar to Jungmin Kang in his 2011 Science & Global Security paper [14]. GIRM was originally proposed by Steve Fetter in 1993 [15] and later further developed by the Pacific Northwest National Laboratory (PNNL) [16–18]. It is a method with which the past plutonium production of a reactor can be estimated without having to rely on accurately knowing its past operation history. One would, however, need to have access to the reactor to take samples at several locations within the core. The idea underlying the method is that the neutron fluence is both directly related to changes in the isotopic compositions of trace impurities in structural components or the moderator and to the cumulative plutonium production in the fuel [16]. For applying GIRM to our simulated ground truth, we proceed in two steps: we first make a local and then a global plutonium estimation.

Local Plutonium Estimation

We deplete a model of a single fuel channel with ONIX [11] and OpenMC [12]. This fuel channel model has periodic boundary conditions in the horizontal x- and y- directions. In vertical direction it has half the extension of the real reactor with a reflective boundary condition on the bottom, making use of the reactor’s symmetry. With the help of this model one can determine the plutonium fluence (cumulative plutonium/cm^3) as a function of the isotopic ratio B-10/B-11. It requires some choice of operational history (discharge burnup, etc.).

By then simulating the ground truth history with the quarter core model shown in Figure 1, one can ‘measure’ the isotopic ratio B-10/B-11 in 100 locations in the core, by reading the isotopic densities from the simulation output. Using the relation derived from the fuel-channel model, the ‘measured’ ratios can be correlated with corresponding plutonium fluence values.

Global Plutonium Estimation

As inspectors will typically not know the precise operational history, we fit a function that is a linear combination of plutonium fluence fields of possible alternative reactor histories to the 100 local plutonium estimates obtained in the local plutonium estimation step, similar to the methodology in Heasler et al. [19]. With the result of this fit we calculate a total plutonium estimate. We repeat this procedure several times, using different plausible reactor histories for the local plutonium estimation and using all possible combinations of fluence fields in the global fit function. For a detailed description of the procedure see [20]. Finally, we average over the results, which we can compare to the ground truth of our core depletion simulation.

Our total plutonium estimate based on the estimation procedure described above is 32.83 ∓ 0.65 kg. It differs by 0.20 kg or 0.62% from our simulated truth of 32.63 kg. In Jung and Göttsche [21], the main source of uncertainty in GIRM are reactor operating parameters. These uncertainties are accounted for in our estimate by averaging over the results we obtain assuming different operational histories in the local estimation step.

4. NEW STATISTICAL APPROACH TO NUCLEAER ARCHAEOLOGY

Although the established GIRM methodology is successful at estimating the amount of plutonium produced in a nuclear reactor, more information about the reactor can be obtained with a new, statistical approach to nuclear archaeology. This approach treats nuclear archaeology as an inverse problem with isotopic ratio measurements as evidence used to reconstruct the operating parameters of a reactor (see Figure 2). Such an inverse problem is solvable with a method called Bayesian inference [22].

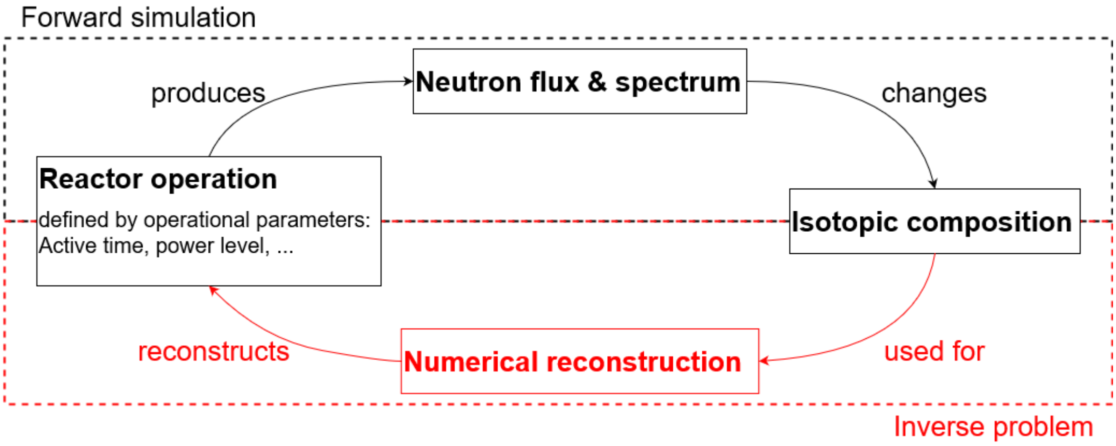

Figure 2: Illustration of the statistical approach to nuclear archaeology

Bayesian inference is a method of statistical inference that uses Bayes' theorem

$$P(A|B) \propto P(B|A)\, P(A)$$

It can be solved using Markov Chain Monte Carlo (MCMC) to approximate the probability distribution of the inference parameters (A) given some evidence (B). Pre-existing beliefs about the parameters are considered in the prior distribution (P(A)), and the likelihood of measuring some evidence given specific parameters is P(B|A). The MCMC algorithm samples parameters and compares the predicted evidence to the measured evidence. By the frequent repetition of this process, the posterior distribution of the parameters is reconstructed.

In this paper, the reconstructed inference parameters are reactor operating parameters, and the evidence is isotopic ratios. Typically, reactor physics Monte Carlo simulations are used to calculate the isotopic composition of materials in reactors. However, such simulations have an untenably high computational cost if repeated frequently. Instead, we use Gaussian process regression (GPR) to create surrogate models that calculate the isotopic ratios much cheaper than Monte Carlo simulations. Figueroa and Göttsche [23] showed that GPR is a suitable technique for creating surrogate models of nuclear waste compositions.

To use the Bayesian inference framework effectively, one needs to select suitable isotopic ratios as evidence. In [24], a combination of global sensitivity analysis and approximate Bayes computation produced good results. We used a similar approach in this work.

In the following, we showcase two applications of the Bayesian inference framework for reconstructing reactor operating parameters. Similar to GIRM, the first approach uses isotopic ratios in the moderator material as evidence. The second uses the composition of high-level reprocessing waste as evidence. Both examples reconstruct the scenario described earlier in Section 2. We simulated this scenario in an infinite lattice fuel channel model to obtain values for the respective isotopic ratios as fictional measured evidence. We assume inspectors know the start and end dates of the reactor operations with an accuracy of plus/minus 6 months for each date. This knowledge, combined with an estimated range of the thermal power (5-25 MW), defines uniform prior distributions for the Bayesian framework. We furthermore assume a blanket uncertainty of 3% on ratio values within the framework.

4.1. Reactor core samples

In a first implementation of our new analysis approach we analyze fictional samples taken from the reactor's graphite moderator. An in-depth explanation of the underlying analysis methodology has been presented previously and can be found in [25] – here we will focus instead on aspects and results specific to the application to the Yongbyon reactor and our ground-truth scenario.

To enable the analysis many isotopic ratios can be be taken into consideration. We therefore extended the description of the moderator material in our reactor model to include all trace elements typically found in graphite of the expected production quality. We based the element list on impurities found in measurements of graphite moderator from the British Trawsfynydd reactor [26], which is similar in design to the Yongbyon reactor.

For this analysis we focused on 5 parameters: the lengths of the two batches' irradiation periods, the average power of the reactor for each of the two batches, and the time passed since the reactor was last active. Figure 3: Posterior distributions for the operating times and power levels of the two batches, as well as the time passed since the reactor was last active, obtained through reconstruction based on the composition of the moderator material. Figure 3 shows the results of the reconstruction for our ground-truth scenario. The reconstruction yields acceptable results for all parameters, however with varying degrees of uncertainty. Reconstruction of the power level of the second, more recent batch is significantly more accurate than for the first one. Reconstruction of the time since the reactor was last operational can be achieved with high precision, while the two operating times show much larger uncertainties.

4.2. High-level reprocessing waste

Spent nuclear fuel is composed of many different nuclides that result from the various nuclear reactions during reactor operation. Characterizing the fuel to determine the fuel burnup is a well-established practice that typically uses only a few specific nuclides (e.g. isotopes of Pu, Nd and Cs [27]). This method only works for fuel elements of a single batch. High-level waste, which is created during one or more reprocessing campaigns, typically contains mixtures of nuclear material of different origins.

Applying the Bayesian framework to the synthetic scenario, we selected four parameters for reconstruction: average burnup and cooling time for each of the two batches of fuel that comprise the mixture of high-level waste. Figure 4 shows the resulting posterior probability distributions. The burnup parameters of both batches are reconstructed well. The marginal distribution, indicated by the blue line, shows a clear peak around the burnup values of the simulated truth. In contrast, the posterior of the cooling time parameter looks as flat as the uniform prior distribution, which indicates that the selected isotopes are not sensitive enough to resolve the cooling time at such high precision. It is possible that further optimizing the isotope selection process will yield better results for the cooling time. Figueroa and Göttsche [24, 28] have shown that the cooling time can be reconstructed in higher burnup scenarios.

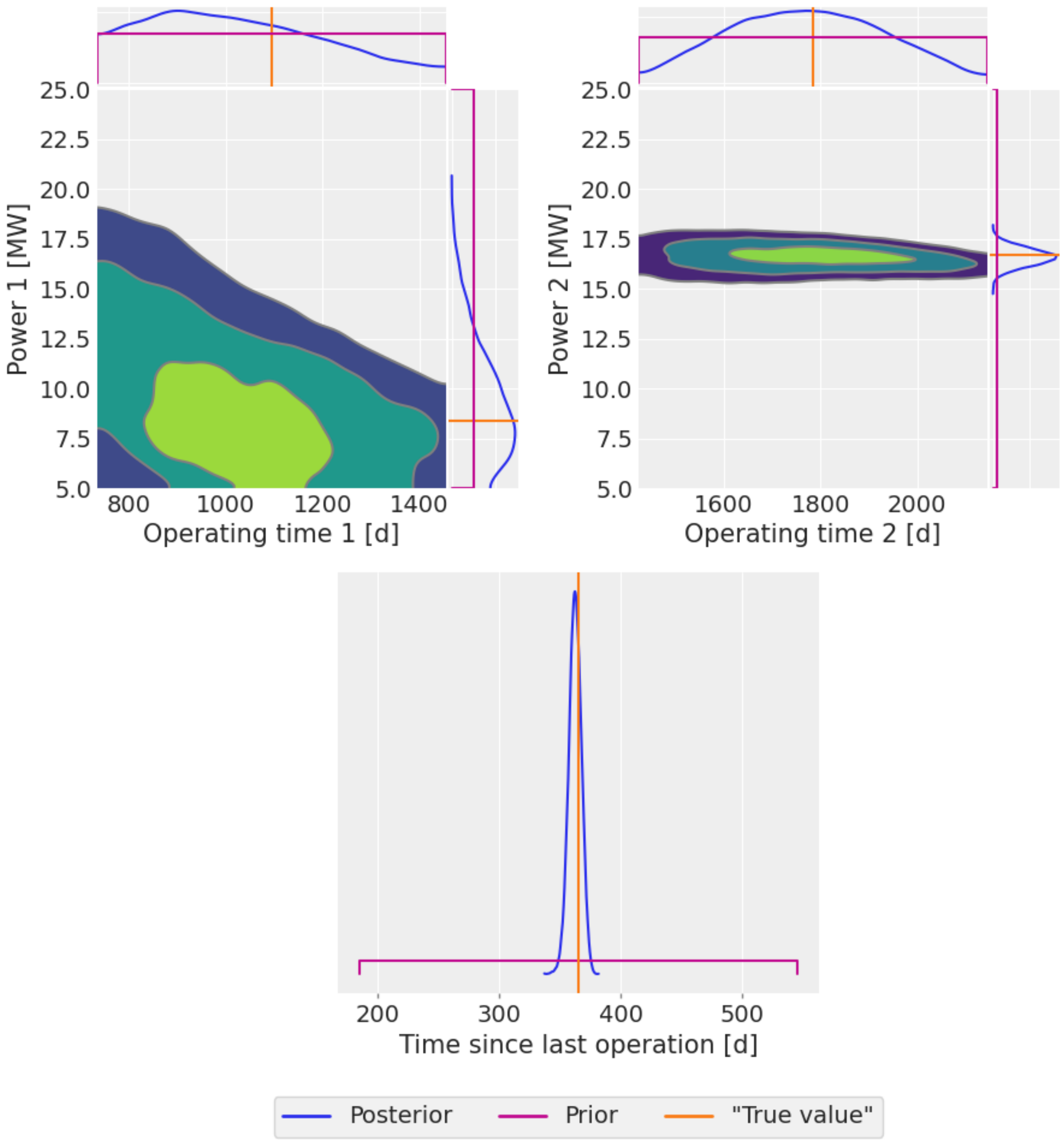

Figure 3: Posterior distributions for the operating times and power levels of the two batches, as well as the time passed since the reactor was last active, obtained through reconstruction based on the composition of the moderator material. The filled areas in the heat maps denote the 1-, 2- and 3-σ sigma regions.

1. CONCLUSION

Nuclear archaeology provides a toolbox to verify initial declaration in a time-efficient manner. As we have shown, sophisticated techniques are being developed to test declarations using various complementary forensic approaches, combined with statistical analyses. GIRM has been validated [18], the Bayesian inference approach using reprocessing waste to some extent as well [24]. The Bayesian approach using graphite measurements is yet to be validated using experimental data. Both latter methods require further study, e.g. regarding a robust choice of the likelihood uncertainty and an optimal isotopic ratio selection.

While it may not be possible to independently reconstruct all information contained in a declaration, nuclear archaeology can be an excellent tool to build confidence nevertheless, as it will be difficult to 'design' a false initial declaration that is consistent with all the measurements.

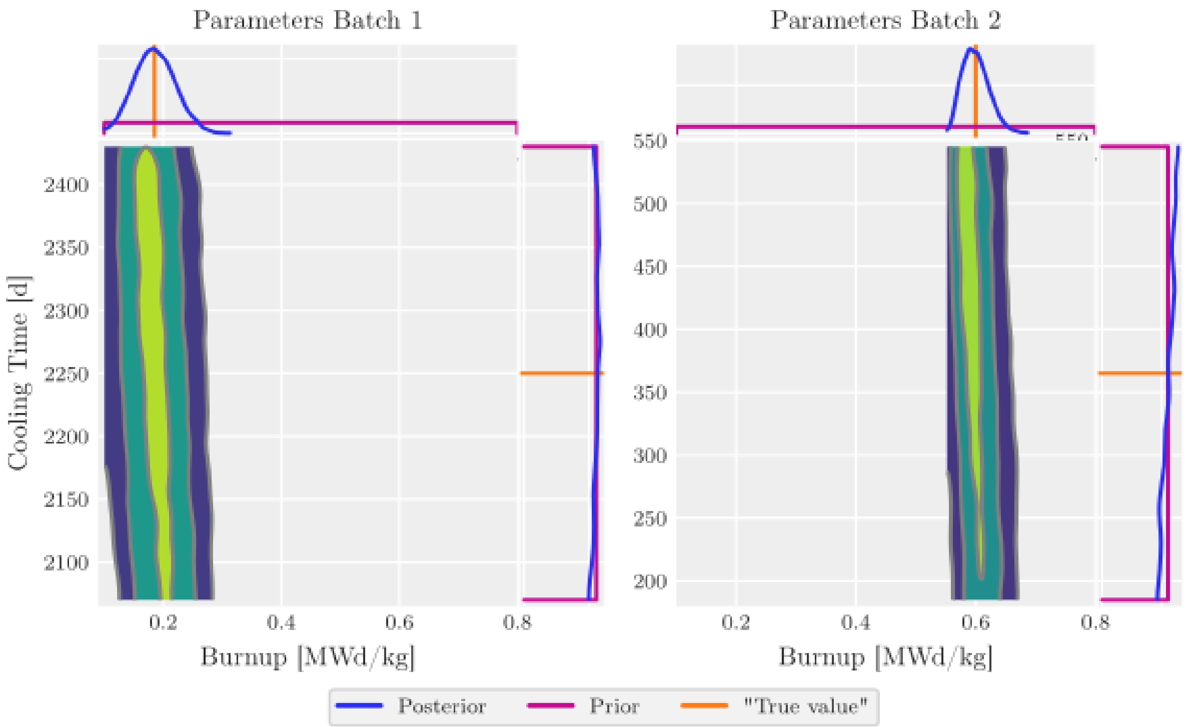

Figure 4: Posterior distributions of the Burnup and Cooling Time parameters of the two batches of fuel, as reconstructed from the composition of the high-level waste. The marginal distributions of each parameter are shown on the sides of each plot.The filled areas in the heat maps denote the 1-, 2- and 3-σ sigma regions.

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